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AIR QUALITY CRITERIA FOR SULFUR OXIDES

National Center for Air Pollution Control
Washington, D. C.

March 1967

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U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
PUBLIC HEALTH SERVICE

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AIR QUALITY CRITERIA
FOR
SULFUR OXIDES

U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
PUBLIC HEALTH SERVICE
BUREAU OF DISEASE PREVENTION AND ENVIRONMENTAL CONTROL
NATIONAL CENTER FOR AIR POLLUTION CONTROL
WASHINGTON, D.C.
MARCH 1967

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FOREWORD

The Clean Air Act of 1963 gave the Department of Health, Education, and Welfare authority to publish air quality criteria as guides for municipal, State, and interstate air pollution control authorities.

These criteria establish the levels of concentration at which individual pollutants begin to harm our health and foul our environment. They set the levels we must aim for in our drive for clean, breathable air.

This volume covers air quality criteria for sulfur oxides. It is the first in a series to be issued by the Department as guideposts for action on the major pollutants in our Nation's atmosphere.

Secretary

PREFACE

The air that we breathe, particularly the air that we breathe in our cities, contains not only nitrogen, oxygen, and carbon dioxide, but a host of other gases and particles, some of which are of natural origin but most of which are wastes from our civilization. The quantities of these other gases and particles relative to the quantities of nitrogen, oxygen, and carbon dioxide are minor, but their effects on human health and welfare may be considerable.

Most gases and particles that pollute the atmosphere are emitted directly from man's activities. Some of these may react with each other or with oxygen or water in the air to form new substances. Some of the newly formed substances may react further to form additional substances, and the extent of all these atmospheric reactions is affected by temperature, relative humidity, sunlight, and the concentrations of the substances. The air, then, at any one time and any one place may very well have unique characteristics. Present technology is not available for anyone to define air quality in all its ramifications, although certain of its characteristics have been found to have a recognizable relationship with effects.

The widespread and growing evidence of the adverse effects of air pollution demands that protective controls be applied now, and if these controls are to be realistically applied, some measure of air quality must be developed.

Recognizing the need for yardsticks of air quality, the Congress in Public Law 88-206, section 103 (c) (2), called upon the Secretary of Health, Education, and Welfare to publish criteria based on the latest scientific knowledge of the predictable effects of various pollutants in the atmosphere.

Air quality criteria, expressed in terms of pollutant concentration and duration of exposure, describe relationships between air pollutants and effects on health and welfare. Such criteria summarize what is known about the effects of pollutants in the atmosphere to provide a realistic base for selecting air quality standards. To achieve or maintain the selected air quality standards, limitations must be placed on the quantities of pollutants that can be emitted. In determining such pollutant emission limitations, to be regulated under law, several additional factors should be considered: existing air quality, and the technical feasibility, relative costs, and time scheduling of the pollution abatement measures.

This document presents criteria of air quality in terms of the effects empirically obtained and published for various concentrations of one family of pollutants, the sulfur oxides, their acids and acid salts.

The abundant use in this country of the sulfur containing fossil fuels, coal and oil, for power and heat, results in these compounds being among the most commonly occurring of the atmospheric pollutants in the United States.

Air polluted with these compounds can produce a variety of effects. Humans vary in their reactions, but such pollution above certain levels unquestionably has adverse effects on the respiratory system and general health. These effects have been particularly evident in several air pollution disasters. High levels can reduce the visual range and prolong fogs. Low levels lasting for long periods can damage materials. When threshold levels are exceeded, vegetation will suffer varying degrees of acute injury depending on pollutant concentration and duration of exposure. Vegetation may also suffer chronic injury from prolonged exposure to concentrations below the threshold for acute injury. The contribution, then, of the sulfur oxides to the effects of many polluted atmospheres is a significant one, and any definition of air quality must include criteria which relate to the effects associated with these oxides.

Although the criteria presented here index the effects of the oxides of sulfur, these effects do not necessarily, nor in fact actually, derive solely from the presence of sulfur oxides in the atmosphere. They are for the most part the effects observed when various concentrations of sulfur oxides, along with other pollutants have been present in the atmosphere. Because of the interactions between pollutants and the reactions of pollutants with oxygen and with water in the atmosphere, and because of the influence of sunlight and temperature on these reactions, the effects of one atmosphere polluted with oxides of sulfur may be quite different from the effects of another atmosphere similarly polluted with oxides of sulfur.

The criteria presented here, then, are not exact expressions of cause and effect that have been replicated from laboratory to laboratory. Instead, the criteria are useful statements of the effects of the sulfur oxides in the atmosphere derived from a careful evaluation of what has so far been reported. As more studies of these effects expand our knowledge, the criteria will be modified accordingly.

The use of these criteria by State and local governments may vary with individual judgment and with local circumstance. In the Federal Clean Air Acts the American people have expressed through their representatives a strong desire for clean air. Guidelines for the choice of criteria are that the quality of the air be good enough that:

1. the health of even sensitive or susceptible segments of the population would not be adversely affected;
2. concentrations of pollutants would not cause annoyance, such as the sensation of unpleasant tastes or odors;
3. damage to animals, ornamental plants, forests and agricultural crops would not occur;
4. visibility would not be significantly reduced;
5. metals would not be corroded and other materials would not be damaged;

6. fabrics would not be soiled, deteriorated, or their colors affected;

7. natural scenery would not be obscured.

The public has spoken for clean air. The criteria presented here offer guidelines for the abatement of air pollution involving sulfur oxides.

I am pleased to acknowledge that these criteria are the result of the pioneering effort carried out under the able leadership of Vernon G. MacKenzie, Deputy Director, Bureau of Disease Prevention and Environmental Control during his tenure as Chief, Division of Air Pollution.

John T Middleton

John T. Middleton
Director
National Center for Air Pollution Control

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SUMMARY

Physical and Chemical Properties of the Oxides of Sulfur

Sulfur dioxide and sulfur trioxide are easily formed during the combustion of sulfur in fossil fuels. About 25 to 30 parts of sulfur dioxide to each part of sulfur trioxide are formed. The other sulfur oxides are either difficult to form or are unstable so that they are not significant air pollutants. Sulfur dioxide is a relatively stable, non-flammable, non-explosive colorless gas that most people can detect at concentrations from 0.3 to 1 ppm in air. Sulfur dioxide can act as either a reducing agent or as an oxidizing agent, and it can react with materials in the air to form sulfur trioxide, sulfuric acid, and sulfate salts. It is highly soluble in water with which it reacts to form sulfurous acid. Sulfur trioxide reacts very rapidly with water to form sulfuric acid, its normal form in the atmosphere.

Atmospheric Reactions of Sulfur Oxides

Sulfur dioxide is oxidized in the atmosphere by either photochemical or catalytic activation. In laboratory studies sulfur dioxide alone in air is oxidized slowly by sunlight (0.1 to 0.2 percent conversion per hour), but if hydrocarbons and oxides of nitrogen are also present in the mixture, photochemical oxidation of sulfur dioxide proceeds at a faster rate. Sulfur dioxide in fog droplets containing iron or manganese salts may be oxidized at the rate of about 1 percent per minute. In field investigations, rates from 0.1 to 2 percent per minute have been observed in the plume of a coal-fired power plant, and an apparent rate of 2.0 percent per hour was calculated in the open atmosphere. Weight ratios of sulfur dioxide to sulfuric acid in ambient air generally are in the range of 0.05 to 0.20, with the larger ratios being associated with higher relative humidities.

Sources of Oxides of Sulfur in the Atmosphere

Of 23,360,000 tons of sulfur dioxide emitted to the air over the United States in 1963, 60 percent was from coal burning, and about two-thirds of that was emitted as a result of burning coal for power generation. Of the remaining 40 percent of the 23,360,000 tons, 15.8 percent was from combustion of residual oils, 4.9 percent from combustion of other petroleum products, 6.8 percent from refinery operations, 7.4 percent from smelting of ores, and the remainder from coke processing (2.0%), sulfuric acid manufacture (1.9%), coal refuse banks (0.8%), and refuse incineration (0.4%).

Measures of Oxides of Sulfur in the Atmosphere

Sulfur dioxide is most commonly measured by the West-Gaeke or hydrogen peroxide technique or by electroconductivity. The West-Gaeke technique underestimates the true concentration of sulfur dioxide in the presence of ozone or nitrogen dioxide or salts of heavy metals. Some of these interferences may be remedied in newer modifications of the technique. The hydrogen peroxide technique may give either high or low values depending on whether there is an excess of basic or acidic gases or particulates other than sulfur dioxide present in the air. Usually the values obtained by this method are intermediate between those obtained by the West-Gaeke and electroconductivity techniques. Electroconductivity methods may give values 1 to 5 times higher than, but usually less than twice as high as, those obtained by the West-Gaeke technique because with electroconductivity methods other soluble gases yield electrolytes in solution and these electrolytes cause high measurements. Sometimes, however, alkaline gases, such as ammonia, which neutralize the acid, are present to the extent that values below those yielded the West-Gaeke technique result. This discussion of measurement errors should not be construed to mean that the measurements do not have value but rather that factors which cause interference should be considered in selecting a measurement method.

Several methods of measuring sulfuric acid mist have been devised. The most extensively used method is an automatic method that makes use of a high-velocity impactor and electroconductivity.

Other indices of sulfur oxides pollution are sulfate content of either suspended particulate matter or settled dustfall and the lead peroxide candle index of sulfur dioxide activity. There is not a common relationship between lead peroxide candle measurements and other measurements of sulfur dioxide concentrations; however, these measurements have value because they can show the presence of very low concentrations over a period of time.

Concentrations and Variations of Oxides of Sulfur in the Air

Although the indicated concentrations of sulfur dioxide vary somewhat with the measurement method, the data of both the National Air Sampling Network (West-Gaeke measurement in approximately 50 locations) and the Continuous Air Monitoring Program (electroconductivity measurement in 6 large cities) indicate that the average annual sulfur dioxide concentrations in cities of the United States range from near zero to as high as 0.16 ppm. Geographically, the concentrations are higher in the northeastern quarter of the United States, with maximum diurnal concentrations usually occurring around 8 o'clock in the morning. Maximum daily average concentrations during a year are approximately 4 times the annual average. Concentrations over a year are approximately log-normally distributed, regardless of averaging time, with 1 percent of the averages exceeding a concentration of approximately 3 times the annual average and 1 percent of the hourly averages exceeding a concentration of about 4.6 times the annual average; 0.1 percent of the hourly averages exceed a concentration that is approximately 7 times the annual average.

Concentrations around large point sources are more variable; at a given point they are usually less than the minimum detectable level, but momentary peaks exceed 1 ppm. Maximum daily averages may exceed 20 times the annual average, and several times a year, concentrations of from 30 to 160 times the annual average may last from a few minutes to an hour or more.

Sulfuric acid mist concentrations up to 50 $\mu g/m^3$ averaged over 30 minutes have been observed in Los Angeles. In Chicago, during the months of November and December 1964, for the hours 10 a.m. to 4 p.m. an average of 9.2 $\mu g/m^3$ was observed.

The national average concentration of suspended sulfates for the years 1957 to 1960 was 11.8 $\mu g/m^3$. Sulfates may constitute a sizeable proportion of the total dustfall; percentages of 10.8 to 32 percent have been reported.

Effects of Air Polluted with Sulfur Oxides on Visual Range

Although direct measurements are not available, the likely effect of sulfuric acid and sulfate salts on visual range can be estimated from existing data on particle size distribution, refractive indices, and concentrations. Because of changes in particle size, at a given concentration of sulfuric acid mist or sulfate salts, visual range is affected more and more as relative humidity increases. If only sulfuric acid mist were involved, at 50 percent relative humidity the estimated visual range would be about 100 miles at a concentration of 10 $\mu g/m^3$ but only about 1 mile at a concentration of 1,000 $\mu g/m^3$; at 98 percent relative humidity, the estimated visual ranges at the same concentrations would be respectively 10 and 0.10 mile. With a normally associated amount of sulfuric acid mist and other particulate matter present, the estimated visual ranges at 50 percent relative humidity would be about 50 and 0.5 mile at measured sulfur dioxide concentrations 0.01 and 1 ppm respectively; at 98 percent relative humidity the estimated visual ranges at the same concentrations would be about 15 and 0.2 miles respectively.

Effects of Air Polluted with Sulfur Oxides on Acidity of Rainwater and Particulate Matter

Data collected in two United States cities indicate that an increase of 12 $\mu g/m^3$ of sulfate in the suspended particulate matter decreases the pH reading of solutions of the particulate matter by an amount between 0.5 and 1. Measurements of the acidity of rainfall have been reported in England, but not in the United States.

Effects of Air Polluted with Sulfur Oxides on Materials

Atmospheres polluted with sulfur dioxide and its associated acidity have been found to be among the most corrosive to metals of all atmospheres studied, even more corrosive than some marine atmospheres. Although considerable corrosion occurs at annual average sulfur dioxide concentrations below 0.05 ppm, its practical significance has not been given adequate attention. Examples of the effects of corrosion include a one-third reduction in the life of overhead line hardware and more than two-thirds reduction in the life of zinc and aluminum facings used in Pittsburgh between the years 1920 and 1940; the necessary use of more expensive, less corrodible metals in some electrical contacts; and one-third of the annual damage to steel rails in England. Air polluted with oxides of sulfur also damages portland cement, various stones, paper, leather, and textiles. Fading of some dyed fabrics is also enhanced by sulfur dioxide.

Effects of Sulfur Dioxide on Vegetation

Sulfur dioxide can cause either acute or chronic injury to vegetation. The limiting concentration that can be tolerated in the cells is about the same for many diverse species. When this concentration is exceeded, the cells die, leaving a characteristic pattern of interveinal and marginal acute injury. Plants with succulent leaves, such as alfalfa, grains, squash, cotton, grapes, and endive, are most sensitive, whereas those with a heavy waxy or cutinized epidermis, such as citrus, pine, and privet, are least sensitive. Under conditions of maximum susceptibility (high light intensity, high relative humidity, adequate moisture supply, and moderate temperature) alfalfa and equally sensitive plants can be injured by exposure to 0.28 ppm averaged over 24 hours, or to 0.48 ppm for 4 hours, or by any combination of exposure time (t) and concentration (c) in which: $(c-0.24)t = 0.94$.

Chronic injury is characterized by a slow oxidation of sulfite to sulfate in the cells. Sulfate toxicity is a form of chronic injury manifested on the leaf by white or brownish-red turgid areas that result from rupture of some of the cells. Abscission of leaves often occurs at or before this stage.

Few studies have indicated that plants are injured when the sulfur dioxide is insufficient to cause visible leaf injury; however, an exposure of alfalfa to 0.10 ppm for 45 days caused some abscission of older leaves, and the growth of rye grass was definitely retarded in ambient air containing daily averages of sulfur dioxide from 0.05 to 0.20 ppm.

Effects of Sulfuric Acid Mist on Vegetation

Sulfuric acid droplets can settle on dry leaves without causing injury, but when the leaf surface is wet, a spotted injury develops. This type of injury has been reported in the Los Angeles area during periods of heavy air pollution accompanied by fog. Swiss chard and beets were reported to be most typically injured; alfalfa develops a spotted injury; and spinach, which is more uniformly wet by the fog, shows a diffuse type of injury.

Acute Toxicity to Animals of Excessively High Exposures

In 8 to 16-hour exposures, around 1,000 ppm of sulfur dioxide is required to kill mice, rats, rabbits, and guinea pigs. In a sulfuric acid mist several hundred milligrams of sulfuric acid per cubic meter of mist is required to kill mice, rabbits, and rats, but as little as 18 mg/m^3 may kill guinea pigs.

Basic Studies with Sulfur Dioxide

Detached single cells in tissue cultures were killed by 8-hour exposures to 5 ppm sulfur dioxide on 2 consecutive days. Although ciliary movement in isolated trachea ceased after 25 minutes of exposure to 7 ppm; it was usually not affected by single exposures of live animals (rats) to 100 ppm or less, and the effect of sulfur dioxide was not enhanced by simultaneously inhaled carbon particles. After exposure of rats to approximately 10 ppm for 18 or 67 days, ciliary movement was diminished, the mucous blanket was thickened and tenacious, and epithelial crypts were demonstrated in the trachea. No pathological changes were observed in other rats after exposure to 40 ppm for 5 hours a day, 5 days a week, but exposure to 300 to 400 ppm for up to 3 months resulted in increased amounts of mucus and goblet cells and the appearance of positive bacterial cultures. It has been reported that in animals breathing 700 ppm only 5 percent of the sulfur dioxide reaches the bronchi whereas at 0.05 ppm 60 percent of the sulfur dioxide reaches bronchi. The percent retention by animals has been shown to decrease with time of exposure and to increase with decreasing concentrations. In humans breathing 16 ppm, only 2 percent of the sulfur dioxide reached the pharynx; however, 84 percent of the sulfur dioxide was retained. Inhaled sulfur dioxide is distributed to all parts of the body. Sulfur dioxide appears to produce increased airway resistance via nerve pathways that stimulate receptors to produce reflex bronchoconstriction.

Effects on Lung Function of Brief Exposures of Animals to Sulfur Oxides

Increases in airway resistance are detectable in dogs exposed to sulfur dioxide concentrations of 1 ppm or more and in guinea pigs exposed to 2 ppm or more. In guinea pigs 20 to 200 times as much sulfur dioxide (weight per volume of air) as sulfuric acid mist or sulfate salts is required to produce a 50 percent increase in airway resistance. Small-sized particulates more effectively increase airway resistance than large ones in the range 0.04 to 7µ. Simultaneously inhaled particles that do not themselves affect airway resistance do, however, enhance the effects of sulfur dioxide, and when sulfur dioxide is inhaled with particles that do increase airway resistance, the increase in airway resistance is greater than the sum of effects of the particles and gas by themselves. Increased airway resistance caused by exposure to irritant particles or to sulfur dioxide in combination with particles persists for much longer periods after discontinuation of exposure than increased resistance caused by sulfur dioxide alone.

Effects of Prolonged Exposures of Animals to Oxides of Sulfur

Information on prolonged exposure of animals to oxides of sulfur is relatively scarce. Rats exposed to 0.16 ppm sulfur dioxide 4 hours per day for 114 days showed reduced activity of dehydrogenase and cholinesterase and reduced vitamin C content in the intestines, kidneys, and liver. Rats exposed to 0.03 ppm sulfur dioxide 5 hours per day for 166 days showed a reversible reduction in carbonic anhydrase. The lifespan of rats exposed over their lifetime to 1, 2, 4, 8, 16, and 32 ppm of sulfur dioxide was significantly reduced at all concentrations. Rabbits exposed to

70 ppm sulfur dioxide for 3 hours a day for 13 weeks showed some changes in the parenchyma of various organs, increased permeability of vascular walls, decreased activity of free sulfhydryl groups, and, in combination with 169 to 320 ppm of carbon monoxide, decreased succinic dehydrogenase activity.

Guinea pigs exposed to 1 ppm or more of sulfuric acid in a mist developed pathological changes in the respiratory tract after a few days. These changes included edema, inter-alveolar wall thickening, interstitual proliferative processes, and lymphoid cell infiltration of blood vessels and beonchi. At 0.5 mg/m^3, sulfuric acid caused slight lung irritation, but when the exposure atmosphere also contained 0.3 ppm of sulfur dioxide, the pathology was similar to that with higher concentrations of sulfuric acid in a mist.

Prolonged Exposures of Animals to Mixtures Containing Oxides of Sulfur

Exposures of animals to sulfur dioxide, sulfuric acid, and combinations of the two indicate that the effects are additive.

After an 80-day exposure of animals to smoke from anthracite, coke, and bituminous coal (sulfur dioxide concentrations were respectively 1.91, 9.12, and 7.51 ppm and particle counts were respectively $3.12 \times 10^8/m^3$, $3.7 \times 10^8/m^3$, and $4.41 \times 10^9/m^3$), red and white blood cell counts and percent hemoglobin were increased. The increases were greatest in animals exposed to bituminous coal smoke, and these animals also showed evidence of fibrosis, proliferation of the bronchial epithelium, and marked peribronchial lymphoid hyperplasia. No significant pathology was observed in lungs of rats exposed to anthracite or coke smoke. Exposure to bituminous coal smoke containing concentrations of from 0.7 to 1.6 ppm sulfur dioxide and from 8.2 to 262 mg/m^3 of particulate matter did not increase infection from Pneumococcus innoculation but did increase the incidence of areas of normally occurring mucus, pus, and caseous material in the lungs of rats.

Industrial Exposures of Humans to Sulfur Oxides

In reported industrial exposures to 10 to 50 ppm sulfur dioxide, no effects were noted in some cases whereas in others there was evidence of deep lung damage, including fibrosis and emphysema. The studies indicate that less serious effects are produced in low humidity atmospheres polluted primarily by sulfur dioxide and that effects increase in seriousness as the atmospheres become more contaminated with other pollutants, such as particulates and sulfuric acid. The more frequent findings include: abnormal urinary acidity; a tendency to increased fatigue; dyspnea; reduced expiratory flow rates; upper respiratory irritations with consequent alteration in the sense of small and taste; nasopharyngitis and bronchitis; increased incidence of cough and expectoration; and increased incidences and increased durations of certain respiratory diseases from infections, including colds and pneumonia.

Experimental Exposures of Humans to Sulfur Oxides

Inhalation of sulfur dioxide in concentrations of 1 ppm or more may increase respiration and pulse rates. The lowest concentration that can induce bronchoconstriction in healthy subjects appears to be between 1 and 2 ppm, and at a given concentration the effect may be enhanced by the simultaneous inhalation of any of several kinds of particulate matter. Occasionally an individual is found who develops bronchospasm upon exposure to sulfur dioxide, and some persons who have been frequently exposed have developed a sulfur dioxide sensitivity.

Shallower and more rapid breathing was observed in subjects inhaling a 350 $\mu g/m^3$ concentration of sulfuric acid mist. In another experiment with a wide range of concentrations of sulfuric acid mists and various exposure times, but with all concentrations at or above 1 mg/m^3, increased airway resistance was reported. It was also demonstrated that wet mists of lower normality (4N) and larger particle size (1.54μ) were more irritating than dry mists of higher normality (10N) and smaller particle size (0.99μ).

Studies of Sensory Threshold Concentrations

The odor threshold for sensitive persons has been reported to be 0.5 to 0.7 ppm sulfur dioxide; the taste threshold is usually considered to be 0.3 to 1 ppm. The odor threshold for sulfuric acid has been reported to be in the range 0.6 to 0.85 mg/m^3.

Sensitivity of the dark-adapted eye to light is increased by sulfur dioxide concentrations in the range of 0.32 to 0.6 ppm. A negligible increase in light sensitivity early in the exposure period, but not later, was observed with a 0.6 mg/m^3 concentration of sulfuric acid in a mist; in the range of 0.7 to 9.6 mg/m^3 the increase was well defined. A combination of 0.22 ppm sulfur dioxide with a 0.3 mg/m^3 concentration of sulfuric acid mist did not increase light sensitivity, but the combination of 1 ppm sulfur dioxide with a 0.7 mg/m^3 concentration of acid resulted in a light sensitivity increase equal to or greater than the sum of the increases produced by these concentrations of the substances individually.

During 20-second exposures to 0.3 to 1.2 ppm of sulfur dioxide a transient desynchronization of the α-wave in the electroencephalogram occurred; greater concentrations caused desynchronization of the α-wave throughout the exposure. Exposure to sulfuric acid mist at concentrations less than 0.6 mg/m^3 did not cause α-rhythm desynchronization but greater concentrations did. Electrocortical conditioned reflexes developed from exposures to: (1) a 0.6 mg/m^3 concentration of sulfur dioxide; (2) a 0.4 mg/m^3 concentration of sulfuric acid mist; (3) the combination of 0.17 ppm sulfur dioxide with 0.15 mg/m^3 of sulfuric acid; and (4) 0.08 ppm sulfur dioxide with 0.30 mg/m^3 of sulfuric acid.

Optical chronaxy was increased by 0.5 ppm sulfur dioxide or by 750 $\mu g/m^3$ of sulfuric acid mist or the combination of 0.3 ppm sulfur dioxide with 600 $\mu g/m^3$ of sulfuric acid mist.

Clinical-Epidemiological Studies on Humans in Community Atmospheres

Several attempts to find clinically measurable differences in the health of individuals exposed to varying degrees of air pollution have not been successful. However, it has been found that the health of bronchitis patients deteriorates when sulfur dioxide concentrations rise above 0.21 ppm and smoke concentrations at the same time rise above 300 $\mu g/m^3$ It has also been found that signs and symptoms of irritation of the respiratory tract, such as cough, increased mucous secretion, nasal catarrh, and wheezing, are related to pollution by the sulfur oxides. Although quantitative differences of respiratory function associated with air pollution have been small and of doubtful significance in some cases, in several independent studies either Wright peak flow rates have been lower or airway resistance higher in persons living in areas with high sulfur dioxide concentrations.

Epidemiologic Studies of Morbidity within Cities

In London, the correlation coefficients of daily variation in either smoke or sulfur dioxide are about 0.3 with daily variation in hospital admissions for respiration morbidity and about 0.2 with daily variations in admissions for cardiac diseases. Correlation coefficients of similar magnitude have been reported between sulfur dioxide concentrations and daily admissions to hospitals in Los Angeles for treatment of cardiorespiratory diseases.

Bronchitis attack rates may be quadrupled during periods of intense air pollution when sulfur dioxide concentrations average from 0.5 to 1 ppm for 3 or 4 days.

When morbidity rates are compared between areas of the same city with different concentrations of sulfur dioxide, the rates of respiratory and cardiovascular illnesses are usually higher in the more polluted areas.

Epidemiologic Studies of Morbidity between Cities

In only two studies have morbidity rates in different cities and the corresponding air pollution concentrations been compared. In one of these a correlation was found between an indirect index of air pollution (percent of days with visibility less than 100 yards at 9:00 a.m.) and man-years lost as a result of bronchitis deaths of postmen while in service or their early retirement because of bronchitis. In the other study a high correlation coefficient was found between the average concentration of suspended sulfate in the air of five cities and the rate of respiratory illnesses lasting more than 7 days in female employees in these same cities.

Epidemiologic Studies of Morbidity Associated with Air Pollution Episodes

Acute illnesses of epidemic magnitude develop when 24-hour-average sulfur dioxide concentrations are approximately 0.5 ppm with peak hourly averages of 0.75 ppm or more, and when at the same time suspended particulate matter concentrations are 1,000 $\mu g/m^3$ or higher, or Cohs values are 8 or more. Increased hospitalization and out-patient clinic visits during these periods are primarily attributed to cardiorespiratory illnesses. In severe episodes accompanied by fog, nausea and vomiting occur in addition to the usual symptoms of respiratory irritation. Secondary complications frequently develop in individuals of all age groups, but the elderly individuals and the individuals with pre-existing cardiorespiratory disease are especially susceptible. From one study it was determined 10 years after the episode that individuals who became ill during the outbreak subsequently had a less favorable morbidity and mortality experience than those who were not so affected.

Epidemiologic Studies of Mortality within Cities

Although it is difficult to isolate the relative importance of each, increases in deaths in London, from "all causes" and from bronchitis occur within 24 hours after daily average sulfur dioxide concentrations reach about 0.25 ppm and black suspended matter concentrations rise above 750 $\mu g/m^3$. Death rates from bronchitis and from "total respiratory diseases" have also been found to be higher in the more polluted areas of Salford, England, and Nashville, Tennessee, respectively, than in the less polluted areas of these cities. In Salford, the average annual sulfur dioxide concentrations in the areas being compared were 0.15 and 0.08 ppm; in Nashville they were 0.027 and 0.013 ppm (interpreted from lead peroxide candle measurements).

Epidemiologic Studies of Deaths Among Cities

Such studies have only been made in England where it has been found that bronchitis death rates relate strongly to indices of air pollution from domestic coal consumption, lead peroxide candle measurements, and pH of the precipitation. Pneumonia death rates were found to increase from 40 to 60 per 100,000 as sulfate in the dustfall increased from 1.4 to 7 tons/mi^2/mo.

Epidemiologic Studies of Deaths During Air Pollution Episodes

During a 4-day period in London, when sulfur dioxide concentrations averaged 0.57 ppm and the smoke concentration 1.41 mg/m^3, 4,000 excess deaths occurred. Such episodes, though of smaller magnitude, have occurred in Donora, Pennsylvania, New York City, and Detroit, Michigan. Such data indicate that episodes occur in United States cities with annual average sulfur dioxide concentrations of 0.05 ppm or more when peak instantaneous concentrations reach 1 ppm or 1/2-hour average concentrations reach 0.8 ppm during stagnation periods lasting 3 or more days.

SUMMARY TABLES

The effects of sulfur dioxide and associated pollutants on man and animals, vegetation, and materials, and the effects of sulfuric acid mist and associated pollutants are summarized in tables I thru IV on the succeeding pages.

Table I. Summary of Effects of Sulfur Dioxide and Associated Pollutants on Man and Animals

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutants		Source	No.
1	TA[a]	<0.015 avg March 17 to Oct. 26	8 other pollutants measured	Increased hospital admissions for respiratory, vascular and infectious diseases on 1/3 highest days as compared to 1/3 lowest days of sulfur dioxide pollution.	Sterling et al., 1966	299
2	WG[b]	0.01 ppm ann avg (arithmetic ann avg approximately 0.015 ppm)	Cohs 0.831	Cardiovascular morbidity in the 55 yr and older age group twice as high as in areas with 1/2 as much pollution.	Zeidberg et al., 1964	298
3	PbO_2[c]	0.49 mg $SO_3/100\ cm^2$ /day; ann avg (equivalent to 0.027 ppm arithmetic ann avg SO_2 by WG)	Cohs >1.1 dustfall >12 tons/ mi^2/mo	Respiratory disease death rate approximately twice as high in middle socioeconomic class living in areas of indicated or higher concentrations as in areas of lower pollution.	Zeidberg et al., 1963	317

(a) Thomas Autometer
(b) West-Gaeke
(c) lead peroxide candles

Table I. (continued)

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
*4	PbO_2	0.4 mg SO_3/100 cm^2/day; air pollution measured only during Aug. and Sept.	Dustfall 35 tons/mi^2/mo	1-second forced expiratory volume and Wright peak flow rates less, after correcting for age, height, sex, and smoking habits, in persons living in town of indicated concentrations than in those living in town with 0.05 mg SO_3/100 cm^2/day and 10 tons/mi^2/mo dustfall; other factors such as ethnic group may account for differences.	Anderson and Ferris, 1965	283
5	PbO_2	0.65 mg SO_3/100 cm^2/day; 3 mo avg low pollution season. 1.2 mg SO_3/100 cm^2/day ann avg	Dustfall 40-182 tons/mi^2/mo	Decreased Wright peak flow rates increased complaints of nonproductive coughs, mucous membrane irritation, and mucous secretion in school children exposed to indicated concentrations as compared to school children living in control area where dustfall was 18-36 tons/mi^2/mo.	Toyama, 1964	80

* Not plotted on figure I

Table I. (continued)

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
6	PG[d]	0.03 ppm 5 hrs per day for 166 days	None	Reversibly reduced blood carbonic anhydrase activity in exposed rats.	Lobova, 1963	217
7	H_2O_2[e] TA	0.03 ppm ann avg Maximum peak 13.5 ppm; several peaks between 1 and 5 ppm	Cohs 2.7	Increased respiratory illnesses in persons exposed to indicated concentrations but no difference in 1-sec forced expiratory volume as compared to persons living in area with 1/4 as much sulfur dioxide pollution and average Cohs of 1.3.	Bell, 1962	285
8	WG	0.037 ppm ann avg	185 $\mu g/m^3$ suspended particulate	Increased frequencies of cough, sputum, dyspnea and bronchitis as compared to frequencies in areas with 0.028 ppm sulfur dioxide and 90 $\mu g/m^3$ suspended particulates.	Petrilli et al., 1966	300

(d) pure gas
(e) hydrogen peroxide

Table I. (continued)

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
9	TA	0.04-0.10 ppm ann avg	Suspended particulate 193-281 $\mu g/m^3$ ann avg	Increased morbidity in people living in areas with indicated concentrations as compared to similar people living in areas with about 1/2 these pollution levels. Differences in morbidity proportional to differences in pollution between comparable areas.	International Jt. Comm. on Pollution in the Atmosphere in the Detroit River Area, 1960	297
10	TA	0.05 ppm ann avg; 3 day pollution build-up to 1 ppm	Suspended particulates 485 $\mu g/m^3$ avg during 3 days	2-fold increase in infant and cancer mortality. Peak SO_2 concentration 1 ppm.	International Jt. Comm. on Pollution in the Atmosphere in the Detroit River Area, 1960	297
11	NMI[f]	0.08 ppm	300 $\mu g/m^3$ H_2SO_4	Can produce cortical conditioned reflexes in man upon repeated 10-sec exposures prior to light flash.	Bushtueva, 1962	264

(f) no method indicated

Table I. (continued)

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
12	WG TA	0.09 ppm 6 mo winter avg Peak hr 2.9 ppm; peak 4.7 ppm	Dustfall 83 tons/mi^2/mo	Interim finding of increased airway resistance in persons exposed to indicated concentrations as compared to persons living in town with 1/9 as much pollution: other factors such as smoking habits, etc. not accounted for.	Prindle et al, 1963	284
13	PbO_2	2.25 mg SO_3/100 cm^2 (equivalent to 0.11 ppm SO_2)		Bronchitis death rates 50% greater in cities with indicated concentrations than in cities with 0.75 mg SO_3/100 cm^2/day.	Pemberton and Goldberg, 1954	328
14	H_2O_2	0.07-0.25 ppm for 4 days		In 4 hospitals there were increased hospital admissions for cardiorespiratory diseases during New York 1953 episode.	Greenburg et al., 1962	310
15	H_2O_2	0.15 ppm ann avg		Bronchitis attack rate and death rate twice as high in areas of Salford, England, with indicated concentrations as in areas with half as much pollution.	Burn and Pemberton, 1963	294

Table I. (continued)

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
16	PG	0.16 ppm 4 hr per day for 114 days	None	50 percent decrease in spleen dehydrogenase activity of 10 exposed rats as compared to 10 control rats.	Lobova, 1963	217
17	E[g]	>0.20 ppm for 4 days (0.9-1.5 ppm hrly max)		Increased incidence of rhinitis, sore throat, cough, and eye irritation observed among 1,000 people.	McCarroll et al., 1964, 1965	277-281
18	H_2O_2	0.20-0.86 ppm for 3 consecutive days	Cohs 6.42-8.38	Air pollution episode in New York City, 1953; 22 excess death per day for 1 week.	Greenburg et al., 1962	345
19	PG[h]	0.2 ppm for 10 sec at repeated intervals in 1 day		Can produce cortical conditioned reflexes in man by repeated 10-sec exposures with light flash.	Bushtueva, et al., 1962	263
20	H_2O_2	0.21 ppm 24 hr avg	Suspended particulates 300 μg/m^3	Deterioration in the health of patients with bronchitis.	Lawther, 1963	315

(g) electroconductivity (other than Thomas Autometer)
(h) pure gas

Table I. (continued)

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
21	H_2O_2	0.25 ppm 24 hr avg	Suspended particulates 750 μg/m^3	Increased total death rates in London.	Lawther, 1963	315
22	H_2O_2	0.25 ppm 6 mo (winter) avg	Suspended particulates 500 μg/m^3	Bronchitis attack rate and death rate twice as high in areas of Salford, England, with indicated concentrations as in areas with SO_2 at 0.10 ppm and suspended particulates at 350 μg/m^3	Burn and Pemberton, 1963	294
23	PG	0.3 - 1.2 ppm for 20 sec		Caused desynchronization of the α-wave for 2-6 sec in the EEG of exposed subjects: lower concentrations did not.	Bushtueva, 1962	263
24	PG	0.3 ppm for 1-2 weeks	500 μg/m^3 H_2SO_4	Guinea pigs developed fibrosis in the interalveolar septi; H_2SO_4 alone at 500 μg/m^3 produced only slight irritation.	Bushtueva, 1960 Bushtueva, 1962	218 219

Table I. (continued)

		Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
25	PG	0.3-1 ppm		Can be detected by average individual (taste).	Patty, 1963	6
26	PG	0.32 ppm for 15 min		Increased sensitivity to light in the dark-adapted eye.	Dubrovskaya, 1957	259
27	H_2O_2	0.46 ppm avg for 15 days max 1.50 ppm for 4 hrs	7 days with Cohs values of 4 or higher	406 excess deaths estimated among population 45 years of age and over.	Greenburg et al., 1965	346
28	PG	0.5-0.7 ppm for 1 sec		Odor threshold for persons with high sensitivity.	Dubrovskaya, 1957	259
29	H_2O_2	0.5-1.0 ppm 24 hr avg for 2-3 consecutive days		4-fold increase over normal in new bronchitis attacks.	Burn and Pemberton, 1963	294
30	H_2O_2	0.57 ppm 5 day avg; 1.3 ppm max daily avg	smoke 1.41 mg/m^3 5 day avg; 4.46 mg/m^3 max daily avg	Air pollution episode in London, 1952; 2.5-fold increase in cardiorespiratory morbidity; 4,000 excess deaths.	Abercrombie, 1953 Wilkins, 1954	309 335

Table I. (continued)

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
31	NMI	0.5-2 ppm (calculated) for 3-4 days		Air pollution episode in Donora, Pa.	Hemeon, 1955	307
32	NMI	0.7-1.6 ppm for up to 165 days	8.2-262 mg/m^3 bituminous smoke	No increase in incidence of or mortality from, Pneumococcus inoculation but 40% increase in incidence of mucus, pus or caseous material in lungs of rats exposed more than 20 wks.	Vintinner and Baetjer, 1951	225
33	PG	1 ppm for 4.5 min		25% increase in light sensitivity of the dark-adapted eye (avg for 5 subjects).	Bushtueva, 1961	261
34	PG	1 ppm for 10 min		Changes in respiration and pulse rate in 14 healthy men aged 23-58 yr.	Amdur et al., 1953	240
35	PG	1 ppm for 20-40 min		Slight decrease in lung compliance and increased airway resistance in anesthetized dogs under controlled breathing.	Balchum et al., 1959, 1960.	192 197

Table I. (continued)

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
36	PG	1-32 ppm 27 months		Lifespan of rats decreased by 0.08 mo for each increase of 1 ppm (calcualted regression coefficient).	Heyssel et al., 1960 Ball et al., 1960	213 214
37	NMI	1.5-60 ppm for 5 min	Hydrogen peroxide aerosol 0.3 mg/m^3 of 1.8μ & 4.6 MMD. Sulfuric acid aerosol 0.01 to 1.4 mg/m^3	No effect on airway resistance from hydrogen peroxide aerosols; but effects of SO_2 enhanced.	Toyama and Nakamura, 1964	251
38	PG	1.6 ppm for 10 min		Threshold for inducing measurable bronchoconstriction in healthy men.	Tomono, 1961	244
39	PG	1.5-80 ppm for 5 min	Sodium chloride aerosol 0.22μ diam	With increasing SO_2 concentrations airway resistance increases at a faster rate with the aerosol than without it (healthy men).	Toyama, 1962	250

Table I. (continued)

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
40	NMI	1.91 ppm for 23 hours per day for 80 days	Anthracite coal smoke 3.12×10^8 particles per m^3	4 rabbits and 8 rats; increased red and white blood cell counts and increased % hemoglobin. No significant lung pathology detected.	Schnurer, 1937	224
41	PG	2 ppm for 1 hr		20% increase in airway resistance of guinea pigs.	Amdur and Mead, 1955	204
42	PG	2-300 ppm for 1 hr	12 mg/m^3 sodium chloride aerosol 0.04 μ diam	With increasing SO_2 concentrations airway resistance increases at a faster rate with the aerosol than without it (guinea pigs).	Amdur, 1957	206
*43	NMI	2-36 ppm for from 1 mo to 44 yrs		Significantly increased frequency of cough, expectoration, and dyspnea on exertion in pulp-mill workers under 50 years of age.	Skalpe, 1964	233

* Not plotted on figure I

Table I. (continued)

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
44	PG	7 ppm for 2 hr per day over 5.5 to 8.5 mo. (0.6 ppm avg.)		20 rabbits immunized against typhoid either after or during exposure showed a substantial drop in agglutination titers.	Navrotskii, 1959	215
45	NMI	7.5 ppm for 23 hours per day for 80 days	Bituminous coal smoke 4.41×10^9 particles per m^3	4 rabbits and 8 rats developed fibrosis, proliferation of the bronchial epithelium, and marked peribronchial lymphoid hyperplasia.	Schnurer, 1937	224
46	NMI	9.1 ppm for 23 hours per day for 80 days	Coke smoke 3.7×10^8 particles per m^3	No significant pathology in lungs of 4 rabbits or 8 rats; moderate increases in red and white blood cell counts and percent hemoglobin.	Schnurer, 1937	224
47	NMI	< 10 ppm avg for > 10 yr		Normal chest x-rays were found in only 7 of 53 foundry workers; abnormalities included fibrosis, enlarged hilum, emphysema, and cardiac disease.	Viikeri, 1956	234

Table I. (continued)

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
48	PG	10-15 ppm for 30-60 min		Rate of removal of mucus from the upper respiratory tract reduced 10-15% in man.	Cralley, 1942	257
49	PG	10 ppm for 18 days		Thickened and tenacious mucous blanket, reduced mucous flow; reduced ciliary activity in exposed rats.	Dalhamn, 1956	188

Table II. Summary of Effects of Sulfur Dioxide and Associated Pollutants on Vegetation

Item	Measure (a)	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
50	TA	0.009 ppm ann avg; maximum daily avg 0.155 ppm		36% of gardens injured.	Sullivan, 1962	81
51	TA	0.03 ppm (avg Apr-Sept.). Occasional peaks in excess of 0.5 ppm for 5 to 10 hr		81% of pine trees had no cones.	Scheffer and Hedgcock, 1955	100
52	TA	0.03 ppm ann avg measurable concentrations present 10-20% of time; concentrations in excess of 0.25 ppm present 1-4% of time		Marked growth retardation and chlorotic needle color of white pine.	Linzon, 1958	83
*53	TA	Concentrations in excess of 0.5 ppm during 10 hr in one mo; occasional peaks in excess of 2 ppm		Acute injury to various tree species.	Linzon, 1965	82

(a) see footnotes to table I
* not plotted on figure II

Table II. (continued)

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
54	TA	0.033 ppm ann avg; max daily avg 0.60 ppm; peaks up to 5-10 ppm		89% of gardens injured.	Sullivan, 1962	81
55	PG	0.10 ppm for 45 days		Premature abscission of older leaves of alfalfa.	Thomas, 1961	141
*56	H_2O_2	0.05 to 0.20 ppm (daily avg) in ambient air during growing season		Retarded growth; increased rate of senescence; reduced tiller formation, number of leaves, and dry weight of rye grass compared to plants grown in washed air.	Bleasdale, 1959	142
57	PG	0.28 ppm for 24 hr or 0.48 ppm for 4 hr or 1.18 ppm for 1 hr		Traces of leaf destruction appear in alfalfa. Calculated from the formula $(c-0.24)t = 0.94$.	Thomas, 1961	141
		1.5 ppm for 24 hr or 1.9 ppm for 4 hr or 3.5 ppm for 1 hr		50% leaf destruction in alfalfa. Calculated from the formula $(c-1.4)t = 2.1$.		

* not plotted on figure II

Table II, (continued)

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
58	PG	0.5 ppm for 7 hr		Acute injury to leaves of various trees and shrubs.	Scheffer and Hedgcock, 1955	100

Table III. Summary of Effects of Sulfur Dioxide and Associated Pollutants on Materials.

Item	Measure (a)	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
59	PbO_2	0.24 mg SO_3/100 cm^2/day (equivalent to 0.007 ppm SO_2) 2 yr avg		Significant corrosion of steel zinc and copper.	Gibbons, 1959 1961	78 79
60	WG	0.09 ppm ann avg	NO_2, O_3	Severe bleaching of dyed fabrics in exposures to ambient urban air.	Salvin, 1963, 1964 National Air Sampling Network Data, 1962, 1963	138 139 33 34
61	NMI	0.15 to 0.05 ppm ann avg 1920 to 1940	Industrial pollution	Life of overhead line hardware reduced 1/3.	Couy, 1948	131-133
62	NMI	0.15 to 0.05 ppm ann avg 1920 to 1940	Industrial pollution	Life of zinc and aluminum facings reduced from 30 to 40 yr each to respectively 7 and 10 yr.	Gilbert, 1954	134

(a) see footnotes to table I

Table III. (continued)

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfur Dioxide	Other Pollutant		Source	No.
63	NMI	0.15 to 0.05 ppm ann avg 1920 to 1940	2-fold reduction in dustfall over same period	4-fold reduction in corrosion of zinc associated with indicated reductions in pollution.	Tice, 1962; Anderson et al., 1961	128 101
64	PG	0.10 to 0.20 ppm 12 hr avg		Certain paint pigments destroyed.	Holbrow, 1962	122
65	PG	0.10 to 0.20 ppm	Ammonia	Formation of crystalline bloom on painted surfaces.	Holbrow, 1962	122

Table IV. Summary of Effects of Sulfuric Acid Mist, Sulfate Salts, and Associated Pollutants

Item	Measure	Concentration and Exposure: Sulfuric Acid or Salt	Concentration and Exposure: Other Pollutant	Effects in Areas with Indicated Concentrations	References: Source	References: No.
66		13 μg/m^3 ann avg total suspended sulfates	Total suspended particulate matter and its components	Increased incidence of respiratory diseases lasting 7 or more days.	Dohan, 1961, 1960	302 303
67		50 μg/m^3 H_2SO_4	0 μg/m^3 of other suspended particulate matter	Visibility less than 10 mi at 70% relative humidity.	Calculated for this report.	
68		100 μg/m^3 H_2SO_4	0 μg/m^3 of other suspended particulate matter	Visibility less than 5 mi at 70% relative humidity.	Calculated for this report.	
69		150 μg/m^3 H_2SO_4 60 to 10 times per day for 10 sec	0.17 ppm SO_2	Produced cortical conditioned reflexes in human subjects.	Bushtueva, 1962	264

Table IV. (continued)

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfuric Acid or Salt	Other Pollutant		Source	No.
70		250 μg/m^3 zinc ammonium sulfate 0.29μ diameter		20% increase in respiratory resistance of guinea pigs.	Amdur and Corn, 1963	208
71		250 μg/m^3 zinc ammonium sulfate 0.29μ diameter	2.5 ppm SO_2	60% increase in respiratory resistance of guinea pigs.	Amdur and Corn, 1963	208
72		350-500 μg/m^3 H_2SO_4 1μ diameter for 5-15 min exposure		40% increased respiration rate; 15 to 20% decreased respiratory flow rate in human subjects.	Amdur et al., 1952 Morando 1956	245 246
73		400 μg/m^3 19 to 23 times in 1 day for 10 sec		Produced cortical conditioned reflexes in humans.	Bushtueva et at., 1962	263
74		500 μg/m^3 H_2SO_4 for 1-2 wks	0.3 ppm SO_2	Guinea pigs developed interstitial proliferative processes in alveolar walls.	Bushtueva, 1960, 1962	218 219

Table IV. (continued)

Item	Measure	Concentration and Exposure		Effects in Areas with Indicated Concentrations	References	
		Sulfuric Acid or Salt	Other Pollutant		Source	No.
75		600 μg/m^3 H_2SO_4		Human odor and irritation threshold.	Bushtueva, 1957	29
76		700 μg/m^3 H_2SO_4 for 4 min		25% increased sensitivity to light in dark-adapted eye.	Bushtueva, 1957	29
77		700 μg/m^3 H_2SO_4	1 ppm SO_2	60% increased sensitivity to light in dark-adapted eye.	Bushtueva, 1961	261
78		910 μg/m^3 zinc sulfate 0.29μ diameter		40% increase in respiratory resistance of guinea pigs.	Amdur and Corn, 1963	208
79		1000 μg/m^3 ammonium sulfate 0.29μ diameter		30% increase in respiratory resistance of guinea pigs.	Amdur and Corn, 1963	208

CONCLUSIONS

Sulfur dioxide is the common, and probably only, oxide of sulfur in the atmosphere; but sulfuric acid, the hydrated form of sulfur trioxide, and its salts are also always present. Although one part of sulfur trioxide is formed for each 25 to 30 parts of sulfur dioxide during the combustion of fossil fuels, the sulfur trioxide is rapidly converted in the atmosphere to sulfuric acid. Sulfur trioxide, and consequently sulfuric acid are also derived from the oxidation of sulfur dioxide in the atmosphere. The concentrations of sulfur dioxide and sulfuric acid and its salts are, therefore, highly correlated, so that sulfur dioxide, which is commonly measured, can be used as an index of total pollution from these compounds. Since only sulfur dioxide has been measured in most reported investigations of oxides of sulfur pollution, criteria of air quality are expressed in terms of concentrations of this gas.

The observed effects of a measured concentration of sulfur dioxide in ambient air are greater than the effects of the same concentration of pure sulfur dioxide. One reason for the more severe effects is the associated presence of sulfuric acid and sulfate salts. Another reason is the interaction of sulfur dioxide and particulate matter in the air.

Health Effects

Among the significant health effects of sulfur dioxide, sulfuric acid, and sulfate salts, individually and collectively, are bronchoconstriction, increased airway resistance, and increased respiration and pulse rates. The resultant stress provides a reasonable explanation for the increased morbidity and mortality observed among individuals suffering from cardiorespiratory diseases as pollution from sulfur oxides increases.

An important property of sulfur dioxide, sulfuric acid, and sulfate salts, particularly the latter two, is their irritant action on the mucous membranes of the respiratory tract. This irritation, which is associated with thickening of the mucous blanket, increased secretion of mucus, and reduced ciliary activity, further explains the increased morbidity and mortality from cardiorespiratory diseases, including respiratory infections and pneumonia.

Figure I was prepared from data reported in table I, pages xxx to xlii, to illustrate the effects of sulfur oxides pollution on health. Analysis of the data permits identification of four areas of differing health significance. The heavily shaded area represents the duration and concentration of exposures generally associated with increased mortality. The cross-hatched area represents exposures associated with reports of increased morbidity. The stippled area represents exposures which are expected to be associated with adverse health effects. The unshaded area represents exposures that are not considered to have any immediately apparent significance.

The extent of the stippled area is based on the following considerations. Items 34, 37, 38, and 39 on figure I show

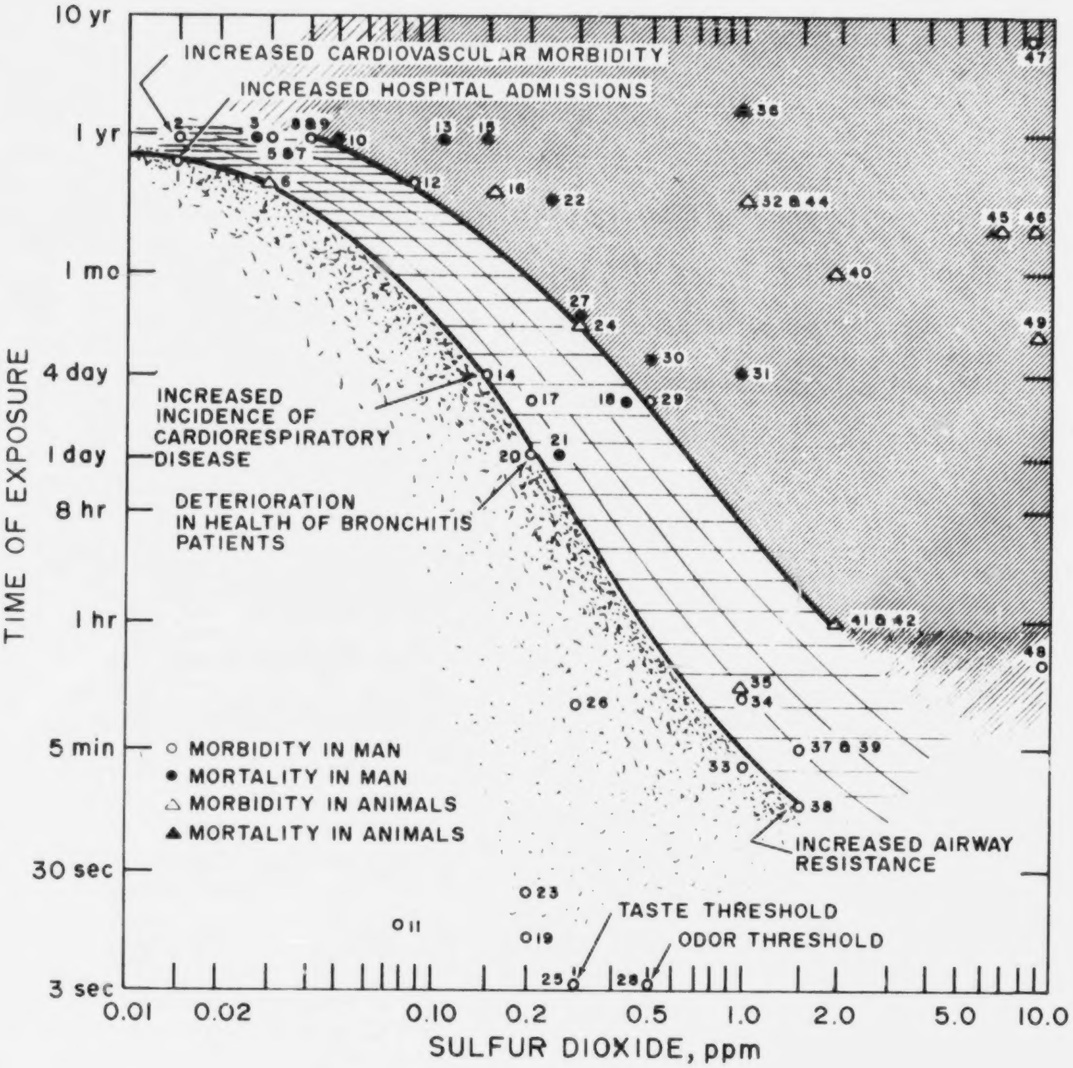

Figure I. Effects of sulfur oxides pollution on health.

Numbers refer to items in column I Table I

▨ Range of concentrations and exposure times in which deaths have been reported in excess of normal expectation.

▦ Range of concentrations and exposure times in which significant health effects have been reported

░ Ranges of concentrations and exposure times in which health effects are suspected cardiorespiratory responses in healthy subjects exposed in the laboratory to 1 to 2 ppm sulfur dioxide for 3 to 10 minutes. It is expected that such responses would be associated with lower sulfur dioxide concentrations in ambient air because: (1) the responses are greater when sulfur dioxide is inhaled simultaneously with particulate matter which is a usual component of ambient air polluted with sulfur oxides; (2) the sulfuric acid mist and sulfate salts which accompany sulfur dioxide in ambient air are also effective in eliciting cardiorespiratory responses; and (3) it is likely that persons with cardiorespiratory conditions would be affected by lower concentrations over comparable times than healthy persons.

Items 14, 17, 20, and 21 show increased cardiorespiratory morbidity and mortality associated with measured sulfur dioxide concentrations in the range of 0.15 to 0.25 ppm averaged over as short a period as 1 to 4 days. However, essentially similar health effects are expected with lower concentrations since cardiorespiratory morbidity has been reported in items 1, 2, 3, 5, 7, 8 and 9 at annual average exposures so low that concentrations of 0.15 to 0.25 ppm averaged over 1 to 4 days would not be likely to occur. Since the effects associated with annual average concentrations are expected to be due to the higher concentrations occurring for shorter periods during the year, increased morbidity and mortality of a degree not detected on a daily basis must occur on days when the average concentrations are less than 0.20 to 0.25 ppm (items 20 and 21).[*]

Hence, it becomes evident that in order to prevent health effects the concentrations and exposure times must be less than the lower limits of the cross-hatched area in figure I. It is not possible to determine from existing data exactly how much lower exposures need to be. However, it is reasonable to assume that the higher concentrations expected for the shorter periods of time, in those urban communities which have the lowest annual average concentration of sulfur dioxide associated with increased morbidity and mortality, provide a basis for estimating the maximal acceptable concentrations for the shorter periods of time. The lower limits of the stippled area were determined from the Continuous Air Monitoring Program data. Since the following concentrations are expected in urban communities with diffuse sources having an annual average concentration of 0.015 ppm sulfur dioxide these sets of ranges of concentrations can be collectively utilized as criteria of acceptable air quality:

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[*] Daily average concentrations of 0.20 to 0.25 ppm are not expected to occur in large urban areas with annual average concentrations below 0.05 ppm. Data collected by the Continuous Air Monitoring Program (CAMP), as reported on pages 34 to 37, provide the means by which expected maximum average concentrations in large urban areas with diffuse sources of oxides of sulfur can be estimated for 24-hour, 1-hour, and 5-minute time periods from any given annual average concentration. These data also provide a means of estimating concentrations that will be exceeded for various percentages of the time. The maximum concentration expected to occur on one day in any one year in the CAMP cities is 3 to 5 times higher than the annual average concentration, and concentrations in excess of a value 2.5 to 4 times the annual average are expected on 3 or 4 days a year. One-hour maximum averages are 8 to 13 times the annual average, and 1 percent of the hourly averages exceed a value 3.5 to 7 times the annual average. Five-minute maximum averages are 8 to 33 times the annual average, and 1 percent of the 5-minute averages exceed a value 3.5 to 9 times the annual average.

Time Period	Maximum	1 Percentile
24-hour average	0.05 - 0.08	0.04 - 0.06
1-hour average	0.12 - 0.20	0.05 - 0.11
5-minute average	0.10 - 0.50	0.05 - 0.14

It might be assumed that the concentrations tabulated above would not be exceeded in any area with an annual average no greater than 0.015 ppm. Special circumstances do occur, however, where atmospheric concentrations are more variable than is usual in large urban areas. For example, due to seasonal space heating requirements, seasonal variation in sulfur oxides emissions may be greater than in CAMP cities. In such places the ratios of short-time averages to annual averages would also be greater, and consequently, more severe effects would be expected. Under such conditions the average concentration during the seasonal or semi-annual period of highest sulfur oxides emissions, rather than the annual average, should be used as the basis for estimating expected concentrations and consequent effects during those periods.

In areas affected primarily by large, single sources, the sulfur dioxide concentrations are usually below the detectable level, but concentrations of considerable magnitude occur briefly from time to time. In such areas, the taste and odor of sulfur dioxide are of prime concern and control measures designed to alleviate these unpleasant sensations should also prevent more serious health effects.

Vegetation Effects

Injury to vegetation by sulfur dioxide has been extensively studied, and the data collected provide relatively precise threshold values for acute injury. Chronic injury from sulfur dioxide exposure and acute injury from sulfuric acid mist have been reported, but exposure concentrations and degree of injury have not been precisely related.

In figure II, the effects on vegetation are plotted from the data in table II, pages xliii to xlv. Available data allow the demarcation of two areas. The shaded area represents exposures that are known to cause injury to vegetation. The unshaded area represents exposures that are below those that cause visible injury. Acute injury to vegetation has occurred in areas with very low annual average concentrations, but where, for short periods of time, emissions from large single sources result in threshold or greater exposures. Since the values required for protection of health are lower than those required for protection of vegetation, it is reasonable to expect that any air quality goals that will adequately protect health will also adequately protect vegetation. In uninhabited areas where vegetation is the prime concern, however, air quality goals should provide some margin of safety to vegetation.

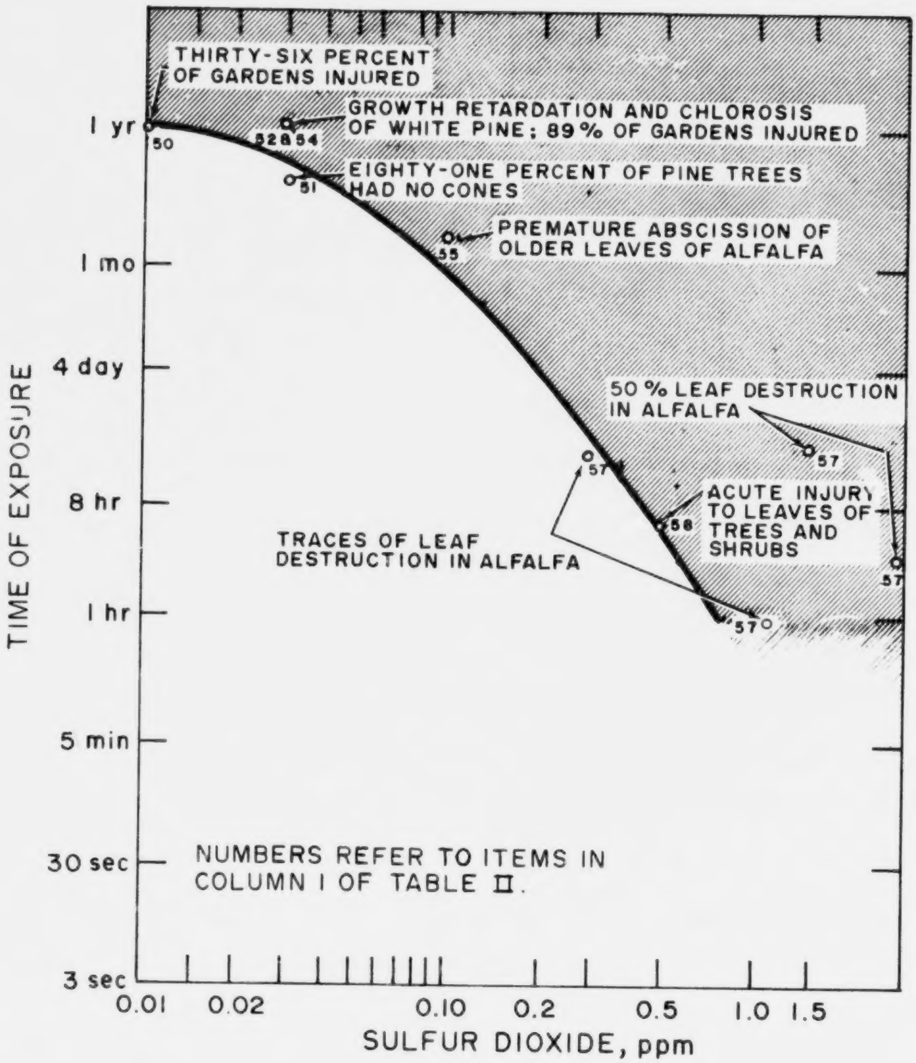

Figure II. Effects of sulfur oxides pollution on vegetation.

▨ Range of concentrations and exposure times in which injury to vegetation has been reported.

☐ Range of concentrations and exposure times of undetermined significance to vegetation.

Visibility Effects

Transmission of light is attenuated by the light-scattering properties of sulfuric acid mist and sulfate salts. Because these compounds are hygroscopic and also because their rate of formation is affected by moisture in the air, their effects on visibility increase with increasing relative humidity. Visibility is reduced by sulfuric acid and sulfate salts, and is further reduced by other particulate matter suspended in the air.

The correlations between sulfur dioxide and sulfuric acid and between sulfur dioxide and other suspended particulate matter were used to estimate the limits of visibility at various sulfur dioxide concentrations. These limits are presented in figure III. The data upon which this graph is based appear on pages 39 to 52.

Observed limits of visibility at given sulfur dioxide concentrations may depart from the calculated values, depending on the concentration of particulate matter and the oxidizing quality of the atmosphere. Nevertheless, the values taken from the graph do provide reasonable guides for estimating the visual ranges to be expected at any given sulfur dioxide concentration.

Materials Effects

The data on corrosion of metals and effects on other materials are summarized in table III, pages xlvi to xlvii. Since the data

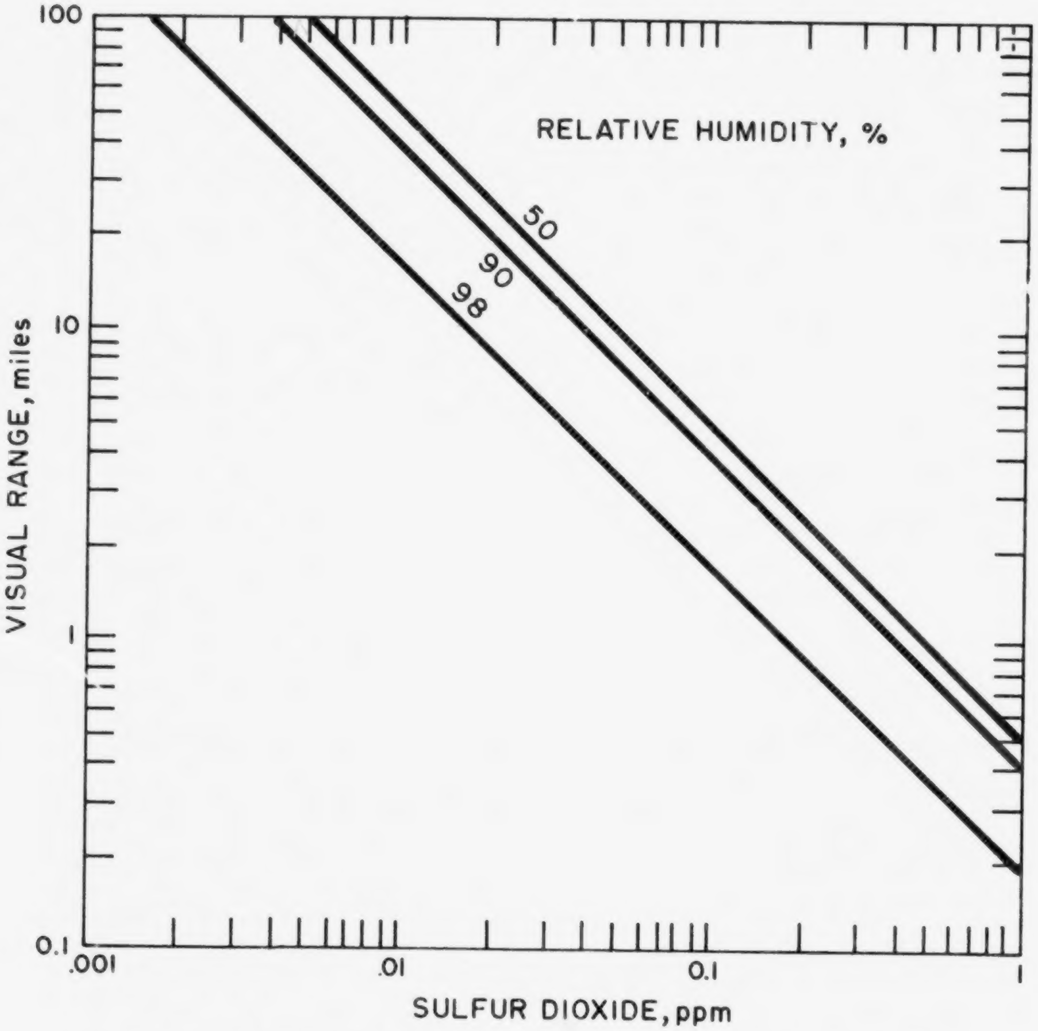

Figure III. Calculated visual range in miles at various sulfur dioxide concentrations.

are largely qualitative, they can be used only in a general way as air quality criteria. The more reactive metals begin to be corroded in moist air when only traces of sulfur dioxide are present and they are severely corroded by the highest reported annual average ambient air concentrations. Reduced corrosion of metals, stone statuary, and buildings, and reduced damage to paper, leather, textiles, and paint will result from any sulfur oxides control.

In summary, if sulfur oxides concentrations are reduced to levels that will protect health, the effects on vegetation will be eliminated, visibility will be appreciably improved, and damage to materials will be markedly reduced.

AIR QUALITY CRITERIA FOR SULFUR OXIDES

INTRODUCTION

"Air Quality Criteria for Sulfur Oxides" has been prepared for the Secretary, Health, Education, and Welfare, by the National Center for Air Pollution Control, Public Health Service, U.S. Department of Health, Education, and Welfare as directed by the Congress in Public Law 88-206.[1]

This document surveys the published scientific knowledge of the occurrence of and the effects due to atmospheric pollution by the sulfur oxides and their acids and acid salts. From this information criteria of air quality based on the effects of sulfur oxides pollution on health and welfare have been prepared for the informational use of municipal, State, and interstate air pollution control agencies.

Preliminary to reviewing the observed effects, this document describes the chemical and physical properties of the sulfur oxides, their characteristic reactions in the atmosphere, sources of sulfur oxides emission in the United States, and the various factors affecting their prevalence in this country, and also reviews various techniques of measuring sulfur oxides. The "effects" portion of the document reviews what presently is reported on the relation between atmospheric sulfur oxides and reduction in visibility, damage to materials and vegetation, and harm to animals and man. This review includes both laboratory and ambient air studies and, for man, includes reports of industrial and experimental exposures and the findings of clinical and epidemiological studies. In the summary of this document, the reported effects of various concentrations of sulfur oxides pollution over a range of concentrations are tabulated in tables I to IV. Two reference lists are provided. In the first list (pages 116-145) the references are in the order in which they appear in the text. The references numbers in the text refer to this list. In the second list (pages 146-176) the references appear in alphabetical order of authors; numbers in parenthesis at the end of each reference refer to the reference number in the first list.

PHYSICAL AND CHEMICAL PROPERTIES OF THE OXIDES OF SULFUR[2,3]

Definition and Occurrence

The oxides of sulfur include sulfur monoxide (SO), sulfur dioxide (SO_2), sulfur trioxide (SO_3), sulfur sesquioxide (S_2O_3), sulfur heptoxide (S_2O_7), and sulfur tetroxide (SO_4). This report also contains information on the acids and acid salts of sulfur dioxide and sulfur trioxide. Sulfur monoxide, sulfur sesquioxide, sulfur heptoxide and sulfur tetroxide are unstable and are difficult to form. Their existence in the atmosphere has not been demonstrated, although it has been suggested that sulfur heptoxide could conceivably exist in some atmospheres as a result of the reaction between sulfur dioxide and ozone.[4]

On the other hand, sulfur dioxide and sulfur trioxide are relatively easily formed. Most fossil fuels, for example, contain sulfur, which burns in air to form sulfur oxides in the ratio of 25 to 30 parts of sulfur dioxide to 1 part of sulfur trioxide. Fossil fuels such as coal and petroleum are burned abundantly in the United States, and sulfur dioxide, sulfur trioxide, and their acids and acid salts are among the most common air pollutants.

Physical Properties of Sulfur Dioxide

Sulfur dioxide is a non-flammable, non-explosive, colorless gas. In concentrations greater than 0.3 to 1 ppm in air it is tasted by most people; in concentrations greater than 3 ppm it has a pungent, irritating odor to most people.[5-7] The gas is highly soluble in water: 11.28 g/100 ml at 20°C, as compared to 0.004, 0.006, 0.003, and 0.169 g/100 ml for oxygen, nitric oxide, carbon monoxide, and carbon dioxide, respectively. The important physical properties of sulfur dioxide are listed in table 1.

Table 1

Physical Constants of Sulfur Dioxide

Molecular weight	64.06
Density (g/l) (gas)	2.927 at 0°C; 1 atm
Specific gravity (liquid)	1.434 at - 10°C
Molecular volume (ml) (liquid)	44
Melting point (°C)	-75.46
Boiling point (°C)	-10.02
Critical temperature (°C)	157.2

Critical pressure (atm)	77.7
Heat of fusion (Kcal/mole)	1.769
Heat of vaporization (Kcal/mole)	5.96
Dielectric constant (Σ)	13.8 at 14.5°C
Viscosity (dyne sec/cm^2)	0.0039 at 0°C
Molecular boiling point constant (°C/1000g)	1.45
Dipole moment (DeBye units)	1.61

Chemical Properties of Sulfur Dioxide and Sulfurous Acid

Sulfur dioxide is a relatively stable gas that can act as a reducing agent or as an oxidizing agent. Of considerable importance to the problem of air pollution is the ability of the gas to react either photochemically or catalytically with materials in the atmosphere to form sulfur trioxide, sulfuric acid, and acid salts. These atmospheric reactions are discussed separately under Atmospheric Reactions of Sulfur Oxides.

In the gaseous state at room temperature sulfur dioxide oxidizes hydrogen sulfide to form elemental sulfur and water, and at elevated temperatures it reacts with the more active metals and amalgams to form oxygen containing anions and sulfides. As a reducing agent the gas reacts very slowly with oxygen at 400°C to produce sulfur trioxide, but catalytic oxidation to sulfur trioxide occurs at temperatures as low as room temperature. Some catalysts that aid this reaction are platinum on a phosphor surface, charcoal, vanadium (V) oxide, graphite, chromiun (III) oxide, iron (III) oxide, and the nitrogen oxides. The nitrogen oxides are used as a catalyst in the Chamber process of manufacturing sulfuric acid from sulfur dioxide.

Ferrous sulfate ($FeSO_4$) catalyzes the reaction of sulfur dioxide directly to sulfuric acid in the presence of oxygen and water.

Some metal oxides oxidize sulfur dioxide directly to sulfate. Aluminum oxide (Al_2O_3), magnesium oxide (MgO), ferric oxide (Fe_2O_3), zinc oxide (ZnO), manganic oxide (Mn_2O_3), cerium oxide (Ce_2O_3), and cupric oxide (CuO) are examples.

Lead peroxide (PbO_2) is an active oxidizing agent and is used in one of the methods for the analysis of sulfur dioxide in air.

Hydrogen peroxide (H_2O_2) is used extensively as an oxidizing agent in the analysis of air samples for sulfur dioxide.

Sulfur dioxide reacts with the halogens. It reacts with chlorine to produce sulfuryl chloride (SO_2Cl_2) and with phosphorous pentachloride to yield thionylchloride ($SOCl_2$). The reaction of sulfur dioxide with iodine is the basis for one of the analytical methods used to measure sulfur dioxide.

Sulfur dioxide reacts with water to form sulfurous acid (H_2SO_3), which is unstable and which exists only in aqueous media. Sulfurous acid is oxidized by the oxygen of the air to sulfuric acid (H_2SO_4) and reacts with iodine to form hydrogen iodide (HI). Sulfurous acid can unite directly with many organic dyes. The

West-Gaeke method for measuring atmospheric sulfur dioxide takes advantage of this property; pararosaniline is used as the organic dye.

Physical and Chemical Properties of Sulfur Trioxide

Sulfur trioxide in ambient air may be emitted directly or may be derived from the oxidation of atmospheric sulfur dioxide. Sulfur trioxide may exist in the air as a gas if the water vapor concentration in the air is low enough, but if sufficient water vapor is present (as there probably always is in ambient air), the gas combines with water to form sulfuric acid droplets, its normal form in the atmosphere. Because of the difficulty of measuring free sulfur trioxide in the air, little is known about how much may be present under various circumstances; presumably it is normally hydrated and present in the unhydrated form only in trace amounts.

Sulfur trioxide reacts as a Lewis acid with a variety of N-containing inorganic ring systems to form addition complexes. It is a strong acid and readily converts basic oxides to sulfates. When sulfates, phosphates, carbonates, perchlorates, and salts of other oxygen acids are reacted with sulfur trioxide, the corresponding anhydride of the oxygen acid is formed. Sulfur trioxide may also act as an oxidizing agent, giving free halogens (except fluorine) with many metal and non-metal halides. Gilbert[8] recently reviewed the numerous and diverse reactions of sulfur trioxide with organic compounds.

ATMOSPHERIC REACTIONS OF SULFUR OXIDES

Laboratory Investigations

Sulfur dioxide is oxidized in the atmosphere by two main processes--photochemical and catalytic. Gerhard and Johnstone[9] determined that the rate of oxidation of sulfur dioxide in 30 percent sulfuric acid drops of 0.3μ diameter in the absence of catalysts is ralatively negligible; however, they pointed out that even in the absence of catalysts the rate of oxidation in a water fog might be faster if rate depends upon the total amount of dissolved sulfur dioxide. These authors estimated that the reaction rate in sunlight was of the order of 0.1 to 0.2 percent conversion per hour, and that the rate was not affected by nitrogen dioxide, sodium chloride nuclei, or relative humidity in the range 30 to 90 percent.

Junge and Ryan[10] studied sulfur dioxide oxidation in solution and found that essentially no oxidation took place in the absence of a catalyst. When iron chloride was used as a catalyst oxidation did take place. The final amount of sulfate formed was relatively independent of the concentration of catalyst but was a linear function of sulfur dioxide concentration. Johnstone and Coughanowr[11] estimated from their study of sulfur dioxide oxidation in small droplets that if manganese sulfate was present as 1μ crystals the oxidation rate in fog droplets would be about 1 percent per minute. Both investigators found that manganese salts were more effective catalysts than iron salts.

Junge and Ryan found that the oxidation of sulfur dioxide essentially stopped when the pH of the water droplets approached 2. They postulated that at least in part this was because of the low solubility of sulfur dioxide in strongly acidic solutions, and they found that if ammonia was present in the air to neutralize the acid as it was formed oxidation of sulfur dioxide continued. Van Den Heuvel and Mason[12] found that for given concentrations of ammonia and sulfur dioxide the mass of sulfate formed was proportional to the product of the surface area of the drops and the time of exposure.

Of primary interest in the photochemical oxidation of sulfur dioxide is the formation of particulate matter in hydrocarbon/nitrogen oxides systems. In the absence of sulfur dioxide little or no particulate matter is formed when atmospheric concentrations of nitrogen dioxide together with atmospheric concentrations of any saturated hydrocarbons are irradiated. However, varying amounts of particulate matter are formed when nitrogen dioxide together with any one of a number of six-carbon and higher acyclic and cyclic olefins and aromatic hydrocarbons (except benzene) are irradiated with sunlight.[13-15] In the absence of nitrogen dioxide, Johnstone and Dev Jain[16] obtained particulate matter when they irradiated sulfur dioxide with n-butane, both at 20mm Hg partial pressure, but Kopczynski and Altshuller[17] were unable to detect any formation of particulate matter when sulfur dioxide at atmospheric concentrations together with atmospheric concentrations of either olefins or paraffins were irradiated. In fact, Renzetti and Doyle[18] and Daintin and Ivin[19] demonstrated that olefins can suppress the production of particulate matter during irradiation of sulfur dioxide in the absence of nitrogen dioxide.

On the other hand, mixtures of olefins, nitrogen dioxide, and sulfur dioxide in the presence of sunlight very definitely form particulate matter, the principal component of which is sulfuric acid.[18,20,21] A primary characteristic of photo-oxidation of sulfur dioxide is the formation of a large number of very small particles with low light-scattering ability. In the absence of other materials the small particles grow slowly and have only a slight light-scattering effect. However, in the presence of olefins and oxides of nitrogen, the particles grow rapidly and light-scattering becomes pronounced. The initial particle diameter may be of the magnitude of 0.02μ; the final particle distribution is dependent upon, among other things, relative humidity. Alkaline material, in general, depresses the growth of particles.

Field Investigations

Gartrell et al.[22] studied the oxidation of sulfur dioxide in coal-burning power plant plumes, using a helicopter to facilitate collection of samples. Soluble sulfates were collected on membrane filters, and sulfur dioxide was collected in hydrogen peroxide. The sulfur trioxide concentration in the stack gas was 15 to 40 ppm and the sulfur dioxide concentration was about 2,200 ppm. Thus, on a weight basis the ratio of sulfuric acid to sulfur dioxide was initially about 0.03. In successive samples collected from the plume, the investigators found that oxidation rates were 0.1 to 2 percent per minute. Increasing rates of oxidation were observed with increasing relative humidity, and the investigators concluded that moisture within the plume or ambient strata was the primary factor affecting the rate of oxidation.

Katz[23] made simultaneous collections of two air samples in the Sudbury, Ontario, nickel-smelting area to determine sulfur dioxide and "total sulfur contaminants," which he interpreted as sulfur dioxide, sulfur trioxide, and sulfuric acid. The total sulfur contaminants sample was collected in a dilute solution of sulfuric acid and hydrogen peroxide and the sulfur dioxide equivalent determined by electroconductivity. The second sample was collected in a starch-iodine solution so that sulfur dioxide could be determined iodometrically. The average ratio of sulfur dioxide to total sulfur contaminants, or net gaseous acid, was highest when the concentration of gases was highest. Katz[24] took this to indicate in an indirect way the extent to which sulfur dioxide is oxidized in the atmosphere, since high concentrations exist for only a short time and concentration is in some measure related to the time the gas has been in the open air.

The change in ratio of sulfur dioxide to total sulfur contaminants may result from processes other than oxidation. For examples, if nitrogen dioxide formed it would be measured as gaseous acid and the measured ratio would be changed and with time the removal of basic substances from the air would change the measured ratio. Further, if the collection apparatus did not efficiently remove particulates and sulfuric acid from the air the data could be misinterpreted.

On the other hand, Katz' results indicate that sulfur dioxide is oxidized in the atmosphere. Katz estimated the approximate time the gas was in the atmosphere, and compared ratios of sulfur dioxide to total sulfur contaminants (expressed as sulfur dioxide equivalent) determined at the beginning and at the end of the period. The ratio decreased from about 95 percent at 1 hour to approximately 65 percent at 12 hours. Since the observations were made in an area relatively free from other contaminants, oxidation of sulfur dioxide is suggested. Analysis of the data presented by Katz showed that the rate of decrease in the ratio of sulfur dioxide to total sulfur contaminants was independent of concentration of contaminants, time of day at which the measurements were made, and ambient temperature.

The following equation was used as a model for the time rate of change of the concentration of sulfur dioxide:

$$\frac{d(SO_2)_t}{dt} = -k(SO_2)_t \quad -p(SO_2)_t$$

where,

$(SO_2)_t$ = the molar concentration of SO_2 at time t

k = the fraction of $(SO_2)_t$ loss per unit time due to oxidation to SO_3, $0 \leq k \leq 1$

p = the fraction of pollutant gases present in the atmosphere replaced per unit time with diluting air, $0 \leq p \leq 1$.

Integrating and applying the boundary condition that,

$$(SO_2)_t = (SO_2)_0 \text{ when } t = 0 \text{ gives the model,}$$

$$(SO_2)_t = (SO_2)_0 \ e^{-(k + p)t}.$$

If $(SO_3)_t$ is the molar concentration of SO_3 at time t, the equation for its time rate of change can be assumed to be:

$$\frac{d(SO_3)_t}{dt} = k(SO_2)_t - p(SO_3)_t$$

where "k" and "p" are the same as defined above.

Substituting for $(SO_2)_t$, integrating, and applying the boundary condition

$$(SO_3)_t = (SO_3)_0 \text{ at } t = 0 \text{ gives}$$

$$(SO_3)_t = (SO_3)_0 \ e^{-pt} + (SO_2)_0 \left[e^{-pt} - e^{-(k + p)t} \right].$$

Hence,

$$R_t = \frac{(SO_2)_t}{(SO_2)_t + (SO_3)_t} = \frac{(SO_2)_0 \ e^{-kt}}{(SO_2)_0 + (SO_3)_0} = R_0 e^{-kt}$$

Taking logs of both sides:

$$\ln R_t = \ln R_0 - kt$$

Showing that under this mathematical model, the ratio of sulfur dioxide to total sulfur contaminants would plot linearly against time on semilogarithmic graph paper.

In the data presented by Katz, the value of k, the instantaneous fractional loss of sulfur dioxide, was found to be 0.021, equivalent to an instantaneous oxidation rate of 2.1 percent per hour or 0.035 percent per minute.

From this oxidation rate it can be calculated that at an initial concentration of 1 ppm sulfur dioxide (2,860 $\mu g/m^3$ at 0°C), and with no dilution, the concentration would be approximately 2,850 $\mu g/m^3$ after 10 minutes, 2,800 μ/m^3 after 1 hour, and 2,300 $\mu g/m^3$ after 10 hours. The corresponding sulfur trioxide as sulfuric acid, would be approximately 15, 90, and 830 $\mu g/m^3$, and the weight ratios of sulfuric acid to sulfur dioxide at the respective times would be approximately 0.005, 0.032, and 0.358.

That much higher rates of oxidation of sulfur dioxide were found by Gartrell and his associates than by Katz may be due in part to more efficient collection of sulfuric acid by the former investigators, and in part due to atmospheric conditions. As noted previously, Gartrell and his associates found that moisture within the plume or ambient air strata is apparently the primary factor affecting rate of oxidation.

There is also the matter of concentration to be considered. Whereas Katz' data indicated that the rate of loss of sulfur dioxide was independent of concentration, Katz' samples, taken in the open air, had sulfur dioxide concentrations of generally less than 2 ppm. Although Gartrell et al. did not indicate concentrations of the contaminants in their plume samples, concentrations in such a plume would be much higher than 1 to 2 ppm, and at the higher concentrations the reaction rate could conceivably be faster.

Coste[25,26] made simultaneous measurements of sulfuric acid and sulfur dioxide in London, England. Sulfur dioxide concentrations ranged from 0.13 to 0.58 ppm (371-1,657 μg/m^3); sulfuric acid concentrations ranged from 4 to 20 μg/m^3. The weight ratio of sulfuric acid to sulfur dioxide was 0.011 at the higher concentration of sulfur dioxide and 0.013 at the lower concentration. The highest ratio of sulfuric acid to sulfur dioxide was 0.023 on a misty day.

Commins[27] reported a maximum sulfur dioxide concentration of 1.47 ppm (4,200 μg/m^3) in London during the period December 2 through 5, 1957. During the same period the maximum concentration of sulfuric acid was 222 μg/m^3. The ratio of maximum sulfuric acid to maximum sulfur dioxide was 0.053. Commins also reported that sulfuric acid could be as much as 10 percent of the total sulfur, a weight ratio of sulfuric acid to sulfur dioxide of 0.167.

Bushtueva[28,29] made an extensive study of the simultaneous presence of sulfur dioxide and sulfuric acid mist in the air. Between August 1953 and January 1954, this investigator collected 198 paired samples for sulfur dioxide and sulfuric acid determination. The data are presented in tables 2 and 3. The sulfuric acid and sulfur dioxide were originally reported in mg/m^3 rather than in μg/m^3 as shown in the tables. The concentrations of sulfur dioxide in ppm have been added for convenience of comparison with other data reported in ppm. These data show that as the sulfur dioxide concentration increases so does the sulfuric acid concentration, although at a slower rate.

Table 2

Correlation Between Concentrations of Sulfuric Acid and Sulfur Dioxide (24-Hour Samples) Bushtueva [28,29]

Concentrations of Sulfur Dioxide		No. of Tests	Average Concentration			Weight Ratio $H_2SO_4:SO_2$
			SO_2		H_2SO_4	
μg/m^3	ppm		ppm	μg/m^3	μg/m^3	
25-100	0.009-0.035	6	0.01	30	12.6	0.420
101-250	0.035-0.088	18	0.06	176	19.6	0.112
251-500	0.088-0.175	38	0.14	387	20.0	0.051
501-750	0.176-0.263	20	0.23	663	31.0	0.045
751-100	0.264-0.350	6	0.30	866	29.0	0.033
1001 and over	0.35 and over	11	0.43	1220	43.0	0.035

Table 3

Correlation Between Concentrations of Sulfur Dioxide and Sulfuric Acid (Single Samples) Bushtueva [28,29]

Concentration of Sulfur Dioxide		No. of Tests	Average Concentration			Weight Ratio $H_2SO_4:SO_2$
			SO_2		H_2SO_4	
μg/m^3	ppm		ppm	μg/m^3	μg/m^3	
up to 250	up to 0.009	15	0.05	128	17.5	0.137
251-750	0.10-0.26	8	0.15	428	41.6	0.097
over 750	over 0.26	2	0.48	1380	326.0	0.235

Bushtueva also studied the effect of wind speed and relative humidity on the concentrations of sulfur dioxide and sulfuric acid. Both the sulfuric acid concentration and the ratio of sulfuric acid to sulfur dioxide were highest during periods of fog, and lowest during periods of precipitation. In the absence of precipitation the ratio of sulfuric acid to sulfur dioxide increased from about 0.045 at 60 percent relative humidity to about 0.090 at 90 percent relative humidity, and to 0.15 at relative humidities above 91 percent. At wind speeds below about 4.5 miles per hour the ratio of sulfuric acid to sulfur dioxide was 0.173, and at wind speeds greater than about 9 miles per hour the ratio was only 0.068. Thus calm days, high humidity, and especially foggy weather were associated with high concentrations of sulfuric acid.

Although they did not study the relationship of sulfuric acid to sulfur dioxide, Mader et al.[30] found that the sulfuric acid concentration in Los Angeles increased as the relative humidity increased.

Chaney[31] studied the relationship between sulfur dioxide (West-Gaeke measurement) and sulfuric acid in the Los Angeles area. During the time of the study sulfur dioxide values were generally very low and sometimes there was no measurable sulfate. The weight ratios of sulfuric acid to sulfur dioxide ranged from 0.037 to 3.0. As stated by the author, the sulfate levels were relatively large compared to the sulfur dioxide concentrations. In the data of the National Air Sampling Network, there is also a large amount of sulfate per unit of sulfur dioxide as measured by the West-Gaeke technique. In the strongly oxidizing atmosphere of Los Angeles, sulfur dioxide may be rather rapidly oxidized. On the other hand, the high ozone and nitrogen dioxide concentrations in Los Angeles may significantly interfere with the West-Gaeke procedure to produce lower than actual sulfur dioxide values. Consequently, oxidation of sulfur dioxide may be more apparent than real.

Thomas[32] used an automatic electric-conductivity-measuring instrument for the simultaneous measurement of sulfur dioxide and sulfuric acid mist during the winter of 1961 in Los Angeles. Values of sulfur dioxide ranging up to 0.21 ppm (600 $\mu g/m^3$) and of sulfuric acid ranging up to 50 $\mu g/m^3$ were observed. Thomas originally reported the data in ppm; but to make them comparable with other data in this report they have been converted to $\mu g/m^3$ and are presented in table 4. The weight ratios of sulfuric acid to sulfur dioxide range from 0.032 to 0.246, these ratios are in the range of values reported by other investigators for other places.

Thomas' data indicate a non-linear relationship between sulfur dioxide and sulfuric acid concentrations. Sulfuric acid increases as sulfur dioxide increases up to some critical value, depending upon the location. Beyond the critical value, sulfuric acid decreases as sulfur dioxide increases. In El Segundo a maximum sulfuric acid concentration of about 25 $\mu g/m^3$ was observed when the sulfur dioxide concentration was between 0.15 and 0.20

Table 4

Relationships of Sulfuric Acid to Sulfur Dioxide in Downtown Los Angeles and El Segundo, Winter of 1963 (Thomas[32])

Date	Sulfur Dioxide		Sulfuric Acid		Weight Ratio
	ppm	μg/m^3	ppm	μg/m^3	$H_2SO_4:SO_2$
			El Segundo		
26 Jan	0.065	185	0.0016	6.4	0.035
6 Feb	0.061	174	0.0031	12.4	0.071
22 Jan	0.062	177	0.0050	20.0	0.112
8 Feb	0.062	177	0.0054	21.6	0.122
10 Feb	0.055	158	0.0050	20.0	0.127
31 Jan	0.120	342	0.0046	18.4	0.054
11 Feb	0.102	291	0.0047	18.8	0.064
28 Jan	0.110	313	0.0055	22.0	0.070
15 Feb	0.125	356	0.0068	27.2	0.076
9 Feb	0.130	371	0.0075	30.0	0.081
30 Jan	0.205	584	0.0048	19.2	0.032
2 Feb	0.194	553	0.0098	39.2	0.071
			Los Angeles		
24 Mar	0.057	162	0.0048	19.2	0.118
21 Mar	0.067	191	0.0070	28.0	0.146
22 Mar	0.063	180	0.0072	28.8	0.160
10 Mar	0.057	162	0.0080	32.0	0.200
9 Mar	0.064	182	0.0092	36.8	0.201
13 Mar	0.065	185	0.0099	39.6	0.214
14 Mar	0.050	143	0.0084	35.2	0.246
22 Mar	0.122	348	0.0072	28.8	0.082
13 Mar	0.122	348	0.0126	50.4	0.145

ppm (425 to 570 $\mu g/m^3$); whereas, in downtown Los Angeles a maximum sulfuric acid concentration of about 30 $\mu g/m^3$ was observed when the sulfur dioxide concentration was between 0.05 and 0.10 ppm (140 to 280 $\mu g/m^3$).

During the past several years the National Air Sampling Network has taken simultaneous measurements of 24 hour average sulfur dioxide and suspended sulfate in various cities.[33-35] Unpublished analyses of these data show that correlation coefficients between sulfur dioxide and suspended sulfate (sulfuric acid and sulfate salts) range between 0.5 and 0.9. Manganese and iron in the suspended particulate matter and relative humidity and temperature were studied as variables that might affect this correlation. As relative humidity and the metals increased so did suspended sulfate. Temperature had no effect on suspended sulfate concentrations.

Thus, the data from field studies essentially agree with data from laboratory investigations; both show that sulfur dioxide can be oxidized to sulfuric acid or an acid salt in the atmosphere. The field studies also show that from evidence taken in a number of geographical locations there is a relationship between sulfur dioxide and sulfuric acid concentrations in the air. The relationship is partly dependent upon the amount of moisture in the air, upon the time the sulfur contaminants have been in the atmosphere, the amount of catalytic particulate matter present in the air, the amount (intensity and duration) of sunlight, the amounts of hydrocarbons and oxides of nitrogen, and the amount of directly reactive and adsorbtive materials in the air, as well as recent precipitation.

SOURCES OF OXIDES OF SULFUR IN THE ATMOSPHERE

Rohrman and Ludwig[36] recently reviewed sources of sulfur dioxide pollution. Sulfur dioxide pollution results primarily from the combustion of fossil fuels, the refining of petroleum, the smelting of ores containing sulfur, the manufacture of sulfuric acid, the burning of refuse, and the burning or smoldering of coal refuse banks. In all of these processes a small amount of sulfur trioxide or sulfuric acid is also emitted.

Specific reviews covering the coal industry, petroleum refineries, fuel oil combustion, burning coal mine refuse banks, sulfuric acid manufacture, and the iron and steel industry have recently been made.[37-44]

Of considerable importance to the meteorological and chemical behavior of sulfur oxides in the atmosphere as well as to their measurement are the kinds of emitters, whether large or small, and whether disperse or point sources. The trend of operations has been away from sulfur dioxide pollution by low-level disperse sources and toward large, point sources, except for diesel trucks using fuels of high-sulfur-content. The large-source emissions contain lower concentrations of polynuclear hydrocarbons and higher concentrations of nitrogen oxides and sulfur trioxide. Also, particulate matter from large sources can be controlled to a greater extent, which means that suspended particulate matter, which interacts with the oxides of sulfur, can be reduced. Further, emissions from large sources are usually emitted from higher stacks, and although this may set the stage for brief early morning fumigations, it reduces average ground level concentrations and the frequency of air pollution episodes.[43-48]

Major sources of sulfur dioxide released to the atmosphere in 1963 as summarized by Rohrman and Ludwig are:

Process	Sulfur Dioxide*	
	Tons	% of Total
Burning of Coal		
Power Generation (211,189,000 tons)	9,580,000	41.0
Other Combustion (112,630,000 tons)	4,449,000	19.0
Sub-Total	14,029,000	60.0
Combustion of Petroleum Products		
Residual Oil	3,703,000	15.9
Other Products	1,114,000	4.8
Sub-Total	4,817,000	20.7
Refinery Operations	1,583,000	6.8
Smelting of Ores	1,735,000	7.4
Coke Processing	462,000	2.0
Sulfuric Acid Manufacture	451,000	1.9
Coal Refuse Banks	183,000	.8
Refuse Incineration	100,000	.4
Total	23,360,000	100.0

* A small amount of this tonnage is converted to sulfur trioxide and sulfuric acid mist before discharge to the atmosphere.

MEASURES OF OXIDES OF SULFUR IN THE ATMOSPHERE

Measures of oxides of sulfur in the atmosphere include various direct indices of sulfur dioxide, measures of the amount of sulfate in suspended particulate matter, measures of the amount of sulfate in settled dustfall, measures of sulfuric acid mist, and the lead peroxide candle index of sulfur dioxide activity.

Direct Indices of Sulfur Dioxide

Katz[49] in 1939 briefly reviewed the methods of indicating sulfur dioxide concentrations which were then in use or had been in use up to that time. The first method for measuring small amounts of sulfur dioxide in the air made use of the bleaching action of sulfur dioxide on a starch-iodine solution. This method was reasonably accurate in the range of 0.8 to 3 ppm sulfur dioxide and was later modified to apply to the range 0.1 to 60 ppm. A method had also been described for measuring the titratable acidity of the air by reacting the air with a water solution of hyperol, the solid compound formed by reacting hydrogen peroxide with urea. Another method involved absorbing sulfur dioxide in 0.1N sodium hydroxide and titrating with iodine. Finally, an automatic instrument, the Thomas Autometer, was developed in which polluted air was drawn through an acidic hydrogen peroxide solution, and the electroconductivity of the final solution measured.

The methods of indicating sulfur dioxide concentrations have recently been reviewed by Jacobs[50] and by Hochheiser.[51] The common methods now in use are the West-Gaeke, hydrogen peroxide, and electroconductivity methods.

The West-Gaeke method has been widely used in the United States in recent years. In this procedure sulfur dioxide is absorbed in 0.1N aqueous sodium tetrachloromercurate to form the non-volatile dichlorosulfitomercurate ion, which is then reacted with formaldehyde and acid bleached pararosaniline to form red-purple pararosaniline methylsulfonic acid. This reaction is specific for sulfur dioxide. The color intensity of the dye, which is proportional to the concentration of sulfur dioxide, is measured at 560 mμ. The method applies to the determination of sulfur dioxide in ambient air in the concentration range from about 0.002 to 5 ppm. In the presence of ozone or nitrogen dioxide, lower concentrations of sulfur dioxide may be indicated than actually exist in the sampled air because the color intensity of the dye is reduced by these compounds. Heavy metals, especially iron salts, oxidize dichlorosulfitomercurate, which also results in low indications of sulfur dioxide. This latter interference can be eliminated by filtration or by including ethylenediamine-tetracetic acid in the absorbing reagent. Hydrogen sulfide precipitates the collecting reagent; such a precipitate must be filtered from the sample prior to determining color intensity. Nitrogen dioxide interference may be eliminated by adding o-toluidine or sulfamic acid subsequent to sample collection.

In the hydrogen peroxide method sampled air is bubbled through a 0.03N hydrogen peroxide solution adjusted to pH 5. Sulfur dioxide present in the sampled air forms sulfuric acid. The total acid collected from the air is then determined by titration with standard alkali. The presence of acidic gases other than sulfur dioxide or reactive acid solids in the air sample gives erroneously high results, whereas the presence of alkaline gases or reactive basic solids gives erroneously low results.

Details for carrying out the procedures of either the West-Gaeke or hydrogen peroxide methods are given by both Jacobs and Hochheiser.

Electroconductivity measuring devices are currently being used in several cities in the United States. These devices are not specific for sulfur dioxide. Electroconductivity is measured in terms of the resistance of the solution between two electrodes immersed in it. Electroconductivity is a property of all ionic solutions, and electroconductivity methods are not specific for any particular compound. Soluble gases that yield electrolytes in solution cause the greatest interference. Any hydrogen halides present are measured. Except near special sources of contamination, however, these gases, compared to sulfur dioxide are seldom present in air in appreciable amounts. Sulfur trioxide gas, if present, would result in a positive interference. Weak acidic gases such as hydrogen sulfide cause practically no interference because of their slight solubility and poor conductivity. Nitrogen dioxide does not interfere appreciably because it is poorly absorbed. If the water is free of bases, the carbon dioxide content of air causes no interference.

Since the particle size of sulfuric acid mist is small (less than 1μ, except when the relative humidity is greater than 85 percent), sulfuric acid mist is not measured appreciably by electroconductivity methods. A special absorber and different operating parameters are required for effective collection of sulfuric acid mist.

Neutral and acidic aerosols such as sodium chloride or sulfuric acid give high results to electroconductivity measurements depending on their solubility, ionization, and the ability of the absorption system to remove them from the airstream, which, in this method, is very poor unless particle size is large.

Alkaline gases such as ammonia interfere with electroconductivity measurements by neutralizing the acid; low results are produced because the transport number of the hydrogen ion is several times greater than that of other cations. Similarly, lime dust or other basic solids, if absorbed, cause comparatively low results for sulfur dioxide.

The methods used in the Soviet Union are of interest. Bushtueva[28] collected sulfur dioxide in a potassium chlorate solution preceded by a glass filter. How the final analysis was made is not stated, but it can be assumed that the procedure was either the nephelometric analysis of barium sulfate or the turbidimetric analysis of lead sulfate.[52,53] Alekseeva and Samorodiva[54] published a fuchsin-formaldehyde procedure in which sulfur dioxide is collecte in glycerol-water solution of 0.01N sodium hydroxide. This method is essentially the same as the fuchsin-formaldehyde method Hochheiser reviewed. Lyubimov[55] reported on the development of a continuous monitoring instrument in which sulfur dioxide is absorbed in a solution of barium chloride. Light transmission through the resulting turbid solution is continuously recorded and is directly proportional to the sulfur dioxide concentration.

The hydrogen peroxide method is most frequently used in Europe; in the United States the West-Gaeke and electroconductivity methods are most frequently used. There is evidence that hydrochloric acid may contribute significantly to the values obtained by the hydrogen peroxide method in England; in general,this would not hold true in the United States because of the negligible amounts of chloride in the coal used in this country and the corresponding negligible amounts of chloride in the atmosphere.[34,56-58]

The values obtained by the various methods of measuring gaseous sulfur dioxide in ambient air may not always correlate well with one another since in general they do not measure the same thing and since interfering substances are present in varying amounts from time to time and place to place. It is generally assumed that sulfur dioxide is the major constituent in air that will react when any of the methods are used, and this may not be entirely true, as some recent studies show.[59-62] The electroconductivity methods may give values 1 to 5 times those obtained by the West-Gaeke procedure; the hydrogen peroxide method usually gives intermediate values.[59,60,62-63] However, when cities are ranked by average sulfur dioxide concentrations as indicated by several of the methods they usually keep the same positions.[64] Therefore, comparisons of sulfur dioxide pollution with average responses of man, plants, and materials usually show similar relationships among cities regardless of which sulfur dioxide index is used. On the other hand, in evaluating responses of man, plants, and materials to day-to-day or hour-to-hour changes in air pollution, the peculiar characteristics of a particular analytical method may be of considerable importance.

Sulfuric Acid Mist Measurements

Data on atmospheric concentrations of sulfuric acid mist are relatively scarce. Collection of samples reflects the inherent problem of collecting particulate matter suspended in the air. Further, methods of analysis for quantitatively differentiating sulfuric acid mist from sulfate salts or other acids are inadequate. The effects of the mist depend on particle size, the determination of which also presents problems related to the behavior and properties of particles.

According to Commins,[27] total acid in London air was determined in about the year 1930 by absorbing both sulfur dioxide and particulate acid in hydrogen peroxide and subsequently titrating with standard alkali. The results of a simultaneous determination of sulfur dioxide by an iodine method were subtracted from the total acid content to give particulate acid concentrations.

Goodeve[65] discussed impingers and filters for removing liquid droplets from air and mentioned the use of glass filters for determining the amount of sulfuric acid in the air. However, he did not elaborate on the analytical method beyond the collection.

Coste[25] mentioned the difficulty of detecting sulfuric acid in air because the microscopic droplets are not absorbed in water or alkaline solutions. However, his co-worker, Courtier, had obtained sulfuric acid from air by first removing sulfur dioxide with hyperol (urea peroxide), passing the air through sintered glass or cotton wool, and then into an ice cooled flask where the water was condensed, collected, and analyzed for sulfate ion by the barium sulfate technique.

Coste and Courtier[26] further modified the method by using lead peroxide instead of hyperol to remove sulfur dioxide from the air. After the air was passed through a U-tube containing lead peroxide absorbed on pumice chips to remove sulfur dioxide, the air was saturated with water. The air was then cooled to condense the water vapor, and the condensate was tested for sulfate and measured for acidity. When the results of the lead peroxide and hyperol procedures were compared, less sulfuric acid was found by the lead peroxide procedure. The authors demonstrated that this phenomenon was not due to sulfur dioxide being oxidized to sulfuric acid in the hyperol procedure, but was due rather to the fact that only 10 to 20 percent of the hygroscopic nuclei present in the sampled air passed through the lead peroxide absorber, whereas 90 percent passed through the hyperol absorber.

Alekseeva and Bushtueva[66] measured sulfuric acid in air by passing air through a lead peroxide filter to remove sulfur dioxide, collecting the sulfuric acid in distilled water, and analyzing the sulfuric acid by the nephelometric barium sulfate technique. They also described a method for the simultaneous measurement of sulfuric acid and sulfur dioxide in the air. The gas and mist were absorbed in a 0.01N solution of sodium hydroxide in 5 percent aqueous glycerol solution. An aliquot of the collected sample was analyzed colorimetrically for sulfur dioxide by a fuchsin-formaldehyde method; another aliquot was treated with hydrochloric acid to displace sulfur dioxide; and the remaining sulfuric acid aerosol was measured nephelometrically.[67]

Mader et al.[30] devised a system capable of filtering 50 to 60 cubic feet of air in 1 hour through Whatman No. 4 filter paper (1-inch diameter) that had previously been washed with distilled water until the washing had a pH of 7 ± 0.10. The test filter was macerated in 20 ml of distilled water, and the pH of the resulting solution determined. Total acidity was then determined by titration with 0.002N sodium hydroxide to an end point of carbon-dioxide-free distilled water and corrected by a blank determination.

Commins[27] described a method in which sulfuric acid in the air was collected by filtering air through Whatman No. 1 filter paper. The filters were then cut in half and one-half was placed in de-ionized water adjusted with 0.01N sodium tetraborate to pH 7 as indicated by bromothymol blue indicator. The solution in which the half filter had been immersed was then titrated to pH 7 with sodium tetraborate to determine the approximate amount of acid present. This amount of acid was less than the true amount since some of the acid reacted with water-insoluble bases **which were** present in the sample. The true amount was found by adding a known excess of 0.10N sodium tetraborate (at least 0.1 ml more than the amount indicated above) to 1 to 2 ml of the pH 7 solution and then immersing the second half of the filter paper in it and titrating the excess with 0.01N sulfuric acid. Acidic gases did not seem to interfere significantly, but large atmospheric concentrations of either particulate acids or various basic constituents, such as ammonia, would have interfered.

Thomas and Ivie[68] described an automatic method for measuring sulfuric acid aerosol in the presence of sulfur dioxide, that used a high velocity impactor. At suitable intervals (usually about 30 minutes) the impactor was washed with conductivity water, and the electroconductivity of the resulting solution determined. The collection efficiency of the impactor was about 70 percent, but varied with particle size, being more efficient with larger particles. The method has recently been used in Los Angeles[32] and Chicago.[69]

Other Indices of Sulfur Oxides Pollution

Sulfate content of either suspended particulate matter or settled dustfall can be determined by conventional methods.[50,70] Sulfate content of suspended particulate matter has been measured for a number of years by the National Air Sampling Network, but in the United States there is little published information on the sulfate content of dustfall. On the other hand, the sulfate of dustfall in England has been determined routinely, whereas the sulfate content of the suspended particulates has not been.[71,72]

The one measurement that has been used extensively in the United States, Canada, England and Japan is the lead peroxide candle index of sulfur dioxide pollution.[59-61,72-80] In this method a paste made of lead peroxide in a gum tragacanth solution is applied to a cotton gauze wrapped around a glass or porcelain form. This is called a lead peroxide candle. The candle is exposed to the air in a louvered shelter for a period of 1 month; the formed lead sulfate is then determined. The method is not specific for sulfur dioxide since it also measures sulfur trioxide, hydrogen sulfide, and other sulfur-containing compounds capable of forming lead sulfate. The importance of the latter method from the standpoint of air quality criteria is that the measurements derived from it have correlated well with certain biological and materials deterioration data.

Correlations between gaseous indices of sulfur dioxide and measures of other oxides of sulfur may also vary from city to city and time of year to time of year for the same reasons that correlations between gaseous measures of sulfur dioxide vary.[60,62]

Reasonably high correlations seem to exist among measures of suspended sulfate, lead peroxide candle sulfation rates, and sulfate content of dustfall.[74] This is to be expected, since the sulfate in all cases is mostly derived from sulfur dioxide in the atmosphere and because one factor, the sulfate ion, is measured in all cases.

CONCENTRATIONS AND VARIATIONS OF OXIDES OF SULFUR IN THE AIR

Sulfur Dioxide

Notable concentrations of sulfur dioxide occur not only in some urban areas that have many disperse sources and/or large sources, but also in rural areas around large point sources such as smelters, power plants, or oil refineries.

Concentrations around large point sources are more variable than concentrations in urban areas where many disperse sources tend to average out the variations associated with wind speed, wind direction, plume rise, and plume dispersion.

Several studies of concentrations around large point sources have been reported. Concentrations at a given point in such an area are usually less than the minimum detectable level, but high concentrations occur under some atmospheric conditions.

Sullivan[81] reported that in the vicinity of a copper smelter in Port Kembla, N.S.W., Australia, the maximum daily average concentrations were 8 to 17 times the annual average concentration. For example, at a station located 1.5 miles from the smelter, whereas the hydrogen peroxide method indicated an annual average of 0.036 ppm, the maximum daily average was 0.60 ppm. A Thomas Autometer indicated peak concentrations from 1 to 5 ppm on 30 occasions during a 2-month period and a maximum peak of 13.5 ppm.

Linzon[82] reported concentrations in excess of 0.5 ppm for 10 hours during one month with momentary peaks over 2 ppm in an area of an oil refinery. He also reported that at a distance of 8 miles from a smelter, measurable concentrations were present about 20 percent of the time. The average concentration over the period of the study (May 1 to Aug. 30, 1954), including the times when there was no measurable sulfur dioxide, was approximately 0.03 ppm; concentrations in excess of 0.25 ppm occurred 4 percent of the time.[83]

Katz[49] reported average concentrations 15 miles south of the Trail, British Columbia, smelter during the 6-month growing season April through September of 0.032, 0.007, 0.008, 0.012, 0.023, 0.022, and 0.013 ppm for the years 1931 to 1937 inclusive. Concentrations in excess of 0.25 ppm occurred during 140, 8.5, 18, 32.5, 36, 47, and 7 hours respectively during these years, or from 0.2 to 3.5 percent of the time. Maximum concentrations lasting for from 4 to 54 minutes ranged from 0.48 to 1.30 ppm or from 30 to 160 times the respective 6-month averages.

Martin and Barber[84] studied sulfur dioxide concentrations around a 1,000 MW power station burning 430 tons of coal of 1.5 percent sulfur per hour. Sixteen sulfur dioxide recorders were spaced around a ring of radius about 3 to 4 miles centered on the station, i.e. near the zone of calculated maximum ground-level pollution. The maximum 3-minute concentration during the year was about 0.62 ppm, the maximum hourly average was about 0.47 ppm, the maximum daily average was 0.11 ppm and the annual average was about 0.027 ppm. It was estimated that the contribution of the power station to the annual average was about 0.001 to 0.002 ppm.

Concentrations of sulfur dioxide around large coal-fired power plants in the United States have not been widely reported, but the data presented by McCaldin and Bye[59] for Seward, Pennsylvania, which is located about one-half mile from such a plant, indicate similar variation. A Thomas Autometer operated during January through April indicated an average concentration of 0.17 ppm (0.09 by West-Gaeke) with a maximum hourly average of 2.9 ppm and a momentary peak concentration of 4.7 ppm.

These data, which show extremely variable concentrations associated with low annual averages, are important because they also show that the thresholds for taste, odor, sensory responses, respiratory responses, and acute injury to vegetation are frequently exceeded.

The major causes of variation in urban concentrations of sulfur dioxide are associated with geographic place, diurnal time, and season of the year, as well as with total rates of emissions from sources.[62,85-95]

Although the indicated concentrations of sulfur dioxide vary somewhat with the measurement method, the data of both the National Air Sampling Network (West-Gaeke measurement in approximately 50 locations) and the Continuous Air Monitoring Program (electroconductivity measurement in 6 large cities) indicate that the average annual sulfur dioxide concentrations in cities of the United States range from near zero to as high as 0.16 ppm.

In the northeastern quarter of the United States, heating and electrical power requirements, customary fuel, and climatological characteristics all combine to generally cause and allow relatively high concentrations.

Heating and electrical power requirements and atmospheric dispersion phenomena combine to cause and allow relatively high concentrations in the fall and winter months. Relatively large requirements of electricity for indoor air conditioning may be responsible for some increased pollution by sulfur oxides during the summer months in some areas.

There is usually a maximum concentration of sulfur dioxide in the morning around 8 o'clock, and a minimum concentration during the afternoon. However, under certain atmospheric conditions, such as stagnating anticyclones, sulfur dioxide can accumulate for several days.

Examination of the Continuous Air Monitoring Program data indicates that the maximum amount by which a given hourly average may differ from the previous hourly average can occur at any time of day, during any month of the year, and regardless of the magnitude of the previous hourly average. The data reveal numerous accumulations of 0.3 ppm or more in 4 hours or less and they also reveal numerous periods of rather steady accumulation of sulfur dioxide over periods of from 1 to 3 days, culminating in concentrations (as indicated by the CAMP monitoring instruments) as high as or higher than 1 ppm even though it is recognized that the CAMP stations are not all located in sections of their respective metropolitan areas where sulfur dioxide pollution concentrations are highest.

There is never a guarantee that a particular set of stagnating atmospheric conditions will break up before the concentrations become critical, as they have in Donora, New York City, Detroit, and areas in other countries, but the accumulation to critical concentrations may be minimized by considering in air quality criteria the usual variation in urban concentrations of sulfur dioxide.

Zimmer and Larsen[99] studied the variation of sulfur dioxide concentrations over 1 year shown by the data of the Continuous Air Monitoring Program and found that the data were approximately log-normally distributed regardless of the averaging time. Although there was slight variation from city to city, 1 percent of the daily averages exceeded a concentration which was approximately 3 (2.5 to 4) times the annual average; 1 percent of the hourly averages exceeded a concentration which was 4.6 (3.5 to 7) times the annual average; and 0.1 percent of the hourly averages exceeded a concentration which was approximately 7 (5.5 to 9) times the annual average. Maximum daily and hourly concentrations, which are less predictable than calculated percentage points of a distribution, were approximately 4 (3 to 5) and 10 (8 to 13) times the annual average concentration. In the National Air Sampling Network data, the ratio of the maximum daily average in a city to the annual average in the same city is approximately 3 (2 to 7).

High sulfur dioxide concentrations are found many miles from either a major urban area or from a large point source.[46,49,101-102] Some measured concentrations 25 to 30 miles from cities are presented in table 5.

Table 5. Sulfur Dioxide Concentrations in ppm at Various Distances From City Centers in 1936-37 and Comparable Data for City Centers in Recent years

Inclusive Dates	Distance from City Center Miles	Pittsburgh		Philadelphia		Washington		St. Louis		Detroit		Methods and Source
		Avg.	Max.	Avg.	Max.	Avg.	Max.	Avg.	Max.	Avg.	Max.	
Oct 12 to Apr 25 1936-37	0-5	0.093	0.90	0.041	0.35	0.014	0.29	0.261	2.27	0.051	0.37	Iodine (Mellon Institute) variable sampling times of about 1 to several hours Reference 102
	25-30	0.083	0.55	0.010	0.08	0.005	0.15	0.026	0.17	0.010	0.18	
Apr 27 to Oct 15 1937	0-5	0.067	0.72	0.031	0.28	0.019	0.14	0.092	1.28	0.036	0.15	
	25-30	0.007	0.04	0.009	0.04	0.006	0.07	0.015	0.05	0.009	0.04	
Oct-Apr 1959-63	0-5	0.035	0.12	0.094	0.27	0.032	0.10	0.041	0.15	0.007	0.07	West-Gaeke sampling times 24 hours National Air Sampling Network, unpublished data
May-Sept 1959-63	0-5	0.020	0.07	0.047	0.13	0.007	0.03	0.029	0.08	0.005	0.04	
Oct-Apr[c] 1963-64	0-5	- -	- -	0.103	0.46(a) 0.84(b)	0.045	0.22(a) 0.39(b)	0.080	0.26(a) 0.83(b)	- -	- -	Electroconductivity CAMP References 80 and 82
May-Sept 1963-64	0-5	-	-	0.046	0.24 0.53	0.027	0.07 0.22	0.048	0.15 0.73			

(a) 24 hour average
(b) Instantaneous observation
(c) Oct-April CAMP data are for Oct., Nov., Dec., 1964, and for March and April 1964

Suspended Sulfate

Sulfate (sulfuric acid and sulfate salts) in suspended particulate matter has been extensively measured for a number of years by the National Air Sampling Network.[58,70] The maximum 24 hour average concentration observed was 94 $\mu g/m^3$. The national average urban concentration estimated from 2,197 samples collected over the years 1957 to 1960 is 11.8 $\mu g/m^3$. There is considerable geographic variation. The observed average concentrations of 18.8, 15.0, 14.5,and 13.3 $\mu g/m^3$ occur in the mid-Atlantic, mideast, midwest and New England areas, respectively. Lower average concentrations of 10.7, 9.0, 8.7, 7.4, and 5.8 $\mu g/m^3$ have been observed in the southeast, Pacific coast, Great Plains, Gulf south, and Rocky Mountain areas, respectively. As with sulfur dioxide the highest concentrations occur most frequently in the fall and winter months.

Sulfate in Dustfall

The Register of Air Pollution Analyses[71] lists only four organizations which have measured and reported sulfate in dustfall in the United States. This measure may be expressed either as percent sulfate in total dustfall, or more usually, as tons of sulfate per square mile per month. In a southwestern city where 32 collecting stations were used, sulfate averaged 10.8 percent of the total dustfall, or 15.5 tons/mi^2/mo over a 3-month period. In a northwestern city sulfate averaged 27 percent of the total dustfall, or 7.4 tons/mi^2/mo, which is similar to the 32 percent of the total dustfall and 7.8 tons/mi^2/mo observed in a New England town.[74-76,101]

Sulfation Rates of Lead Peroxide Candles

Considerable use has been made of lead peroxide candles in the United States, and usually they have been used in conjunction with other measures of sulfur oxides pollution. Sulfation rates in United States cities have been observed to range from a few tenths of a milligram to about 8 mg of sulfur trioxide per 100 cm^2 of exposed lead peroxide candle surface per day (mg SO_3/ 100 cm^2/day).[59-61,74]

Sulfuric Acid Mist

Only a few attempts have been made to measure sulfuric acid mist in the United States. Concentrations measured in Los Angeles average about 25 μg/m^3, and a high concentration of 50 μg/m^3 has been observed.[30-32] In Chicago, during the months of November and December 1964 the average for the hours 10 a.m. to 4 p.m. was 9.2 μg/m^3.[69]

EFFECTS OF AIR POLLUTED WITH OXIDES OF SULFUR ON VISUAL RANGE, MATERIALS AND VEGETATION

Visual Range

One of the most noticeable physical effects of air pollution is its effect on light transmission as evidenced by reduced visibility. Comprehensive treatments of the subject have been made by Steffens,[103] Middleton,[104] and Robinson.[105]

In the atmosphere light is scattered and in addition is attenuated by absorption. Of the two effects, scattering is the more important effect of sulfur oxides pollution.[106]

The exact contribution that the oxides of sulfur make to the total scattering of light by various atmospheres has not been well studied. The sulfur oxides products that cause scattering are sulfuric acid mist and other sulfate particulates.

Visual range V_2, along a given path is arbitrarily defined as the distance a black box target must be moved from an observer to reduce its contrast with the horizon at the sky to 0.02 and is given by the relation:

$$V_2 = \frac{3.92}{\sigma} = \frac{3.92}{NAE}$$

where,

σ is the attenuation per unit path length

N is the number of particles per unit volume of atmosphere with cross sectional area A

E is the particle extinction coefficient.[105]

The particle extinction coefficient, E, represents the total light scattered and absorbed by a particle divided by the light geometrically incident on the particle. This coefficient depends on the particle's refractive index, its shape, and its size relative to the wavelength of the light and usually is expressed as,

$$\alpha = \pi d/\lambda$$

where,

d is the particle diameter

λ is the wavelength of light in the medium surrounding the particle.

Refractive indices of sulfuric acid in equilibrium with water vapor at various relative humidities are given in table 6 and in table 7 values for E when $\lambda = 0.5\mu$ are given for various concentrations of sulfuric acid mist of various particle sizes.

When particles of different sizes or different refractive indexes are involved, the equation $V_2 = 3.9/NAE$ for visual range must be modified as follows:

$$V_2 = \frac{3.9p}{\Sigma N_i A_i E_i}$$

where:

V_2 is the visibility in standard units with a contrast limen (threshold) of -0.02

i identifies a particle of a given diameter (d) and a given refractive index (RI)

N_i represents the number of i particles per unit volume

A_i represents the cross sectional area of an i particle

E_i is the extinction coefficient of an i particle

p is 1 if V is in the same units as N and A; e.g. V_2 is in meters if N_i is number of particles per cubic meter and A_i is cross sectional area in square meters

but,

p is 62.14×10^{-5} if V_2 is in miles, N_i is in number of particles per cubic meter, and A_i is in square meters

Waller and co-workers[110,111] studied acid droplets in urban air and presented data pertaining to a 39 $\mu g/m^3$ sample of sulfuric

Table 6. Density, Percent Sulfuric Acid, and Refractive Index of Sulfuric Acid Solutions in Equilibrium With Water Vapor at Different Relative Humidities [107-108]

Relative Humidity %	% H_2SO_4	Density	Refractive Index
0	100.00	1.8305	1.440
2	84.41	1.7615	1.434
5	69.44	1.6015	1.421
10	64.45	1.5485	1.414
20	57.76	1.4775	1.407
30	52.45	1.4205	1.399
40	47.71	1.3705	1.393
50	43.10	1.3305	1.387
55	40.75	1.3105	1.384
60	38.35	1.2885	1.381
65	35.80	1.2665	1.378
70	33.09	1.2435	1.374
75	30.14	1.2205	1.370
80	26.79	1.2025	1.366
85	22.88	1.1645	1.362
90	17.91	1.1265	1.356
95	11.08	1.0745	1.347
97.5	7.42	1.0385	1.343
98	4.99	1.0315	1.340

Table 7. Extinction Coefficients for Various Size Droplets of Sulfuric Acid Mist in Equilibrium With Water Vapor at Various Relative Humidities (Interpolated from Reference No. 109)

	PERCENT RELATIVE HUMIDITY										
d*	50	55	60	65	70	75	80	85	90	95	98
0.1	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.01
0.2	0.24	0.24	0.23	0.23	0.22	0.22	0.21	0.21	0.20	0.19	0.18
0.3	0.88	0.87	0.85	0.84	0.81	0.80	0.78	0.76	0.73	0.69	0.66
0.4	1.63	1.60	1.58	1.56	1.53	1.50	1.47	1.44	1.40	1.33	1.28
0.5	2.50	2.42	2.43	2.41	2.36	2.32	2.27	2.23	2.17	2.07	1.99
0.6	3.18	3.13	3.11	3.06	3.02	2.93	2.92	2.88	2.81	2.69	2.60
0.7	3.63	3.58	3.52	3.53	3.50	3.46	3.42	3.39	3.34	3.23	3.16
0.8	3.94	3.92	3.91	3.89	3.87	3.81	3.84	3.82	3.78	3.72	3.67
0.9	4.00	4.00	3.99	3.98	3.97	3.98	3.97	3.97	3.95	3.94	3.92
1.0	3.84	3.85	3.85	3.85	3.86	3.87	3.87	3.87	3.88	3.89	3.89
1.1	3.30	3.33	3.31	3.38	3.42	3.45	3.48	3.52	3.58	3.67	3.73
1.2	2.84	2.93	2.98	3.02	3.53	3.14	3.21	3.27	3.35	3.49	3.59
1.3	2.33	2.50	2.54	2.58	2.65	2.71	2.78	2.84	2.93	3.07	3.16
1.4	2.20	2.23	2.22	2.29	2.33	2.38	2.37	2.46	2.52	2.63	2.70
1.5	1.78	1.85	1.81	1.88	1.90	1.92	1.95	1.99	2.05	2.16	2.27
1.6	1.66	1.61	1.62	1.68	1.71	1.72	1.76	1.79	1.86	1.90	2.08
1.7	1.92	1.91	1.91	1.91	1.90	1.90	1.90	1.90	1.90	1.90	1.90
1.8	2.11	2.12	2.10	2.08	2.04	2.01	1.98	1.94	1.89	1.82	1.75
1.9	2.47	2.42	2.39	2.35	2.30	2.25	2.21	2.16	2.08	1.91	1.88
2.0	2.43	2.40	2.43	2.40	2.32	2.33	2.29	2.26	2.20	2.10	2.03

* diameter in microns

acid mist with a mass median diameter of 0.5μ and a geometric standard deviation of 8. The relative humidity at the time the sample was collected was 85 percent. From the density of sulfuric acid in equilibrium with water vapor at 85 percent relative humidity and the mass distribution of the sample, the number of particles of various sizes was calculated.

If particles of all sizes reach the same equilibrium sulfuric acid concentration at a given relative humidity (table 6), the number of particles of each size at any other relative humidity may be calculated from the relationship:

$$n_{d'_{RH'}} = n_{d_{RH}}$$

where,

$n_{d'_{RH'}}$ is the number of particles of diameter d at a given relative humidity RH which have either increased or decreased in size to diameter d' with a change in relative humidity to RH'

Given the diameter d of particles at a specific relative humidity RH, the diameter d' of particles at relative humidity RH' can be determined from the relationship:

$$d'_{RH'} = \left| \frac{W_{d_{RH}} \quad C_{H_2SO_{4_{RH}}}}{C_{H_2SO_{4_{RH'}}} \quad 1.91 \quad d_{RH}} \right|^{1/3}$$

where,

$W_{d_{RH}}$ = the weight of a particle of diameter d at relative humidity RH

$C_{H_2SO_{4_{RH}}}$ = the equilibrium weight proportion of H_2SO_4 in the droplets at relative humidity RH

$C_{H_2SO_{4_{RH'}}}$ = the equilibrium weight proportion of H_2SO_4 in droplets at relative humidity RH'

1.91 = the ratio of the cube of the diameter of a sphere to its volume

Data from Waller's sample, were used to calculate mass median diameters for various relative humidities (figure 1). The resulting calculated mass median diameters agree well with those observed by Ludwig and Robinson[112] at corresponding relative humidities.

An important corollary to the increase in mass median diameter with increase in relative humidity is that at higher relative humidities there are greater numbers of particles (figure 2) in the size range of 0.1 to 2.0μ, the size range that significantly affects visibility reduction. The consequences are shown in figure 3 where, for example, it can be seen that at 30 $\mu g/m^3$ of sulfuric acid mist the visibility is calculated to be 31 miles at 50 percent relative humidity but only 3.1 miles at 98 percent relative humidity.

Sulfuric acid mist in the atmosphere is infrequently measured. However, since several investigators have reported increasing ratios of sulfuric acid mist to sulfur dioxide with increasing relative humidity, and since correlations between sulfur dioxide

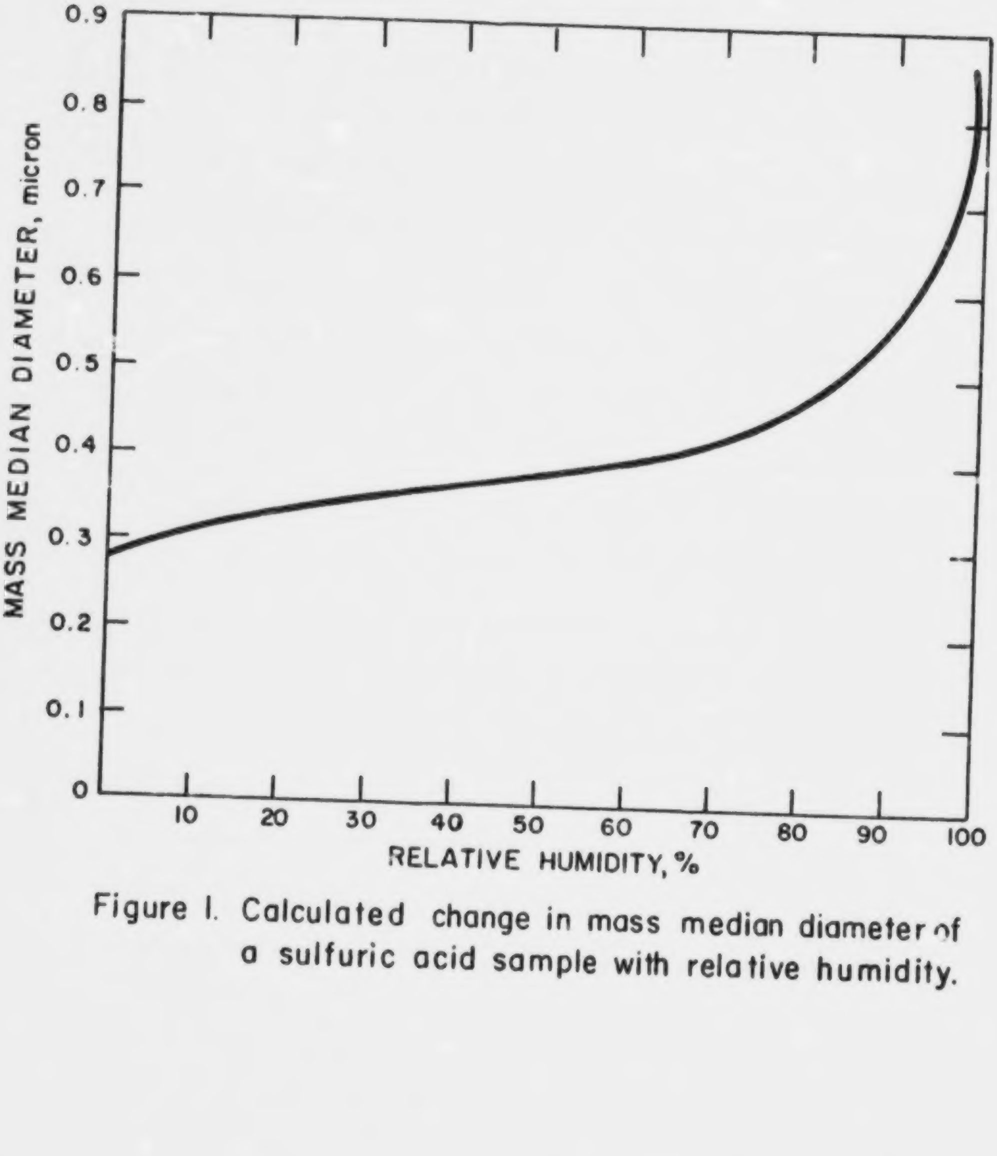

Figure 1. Calculated change in mass median diameter of a sulfuric acid sample with relative humidity.

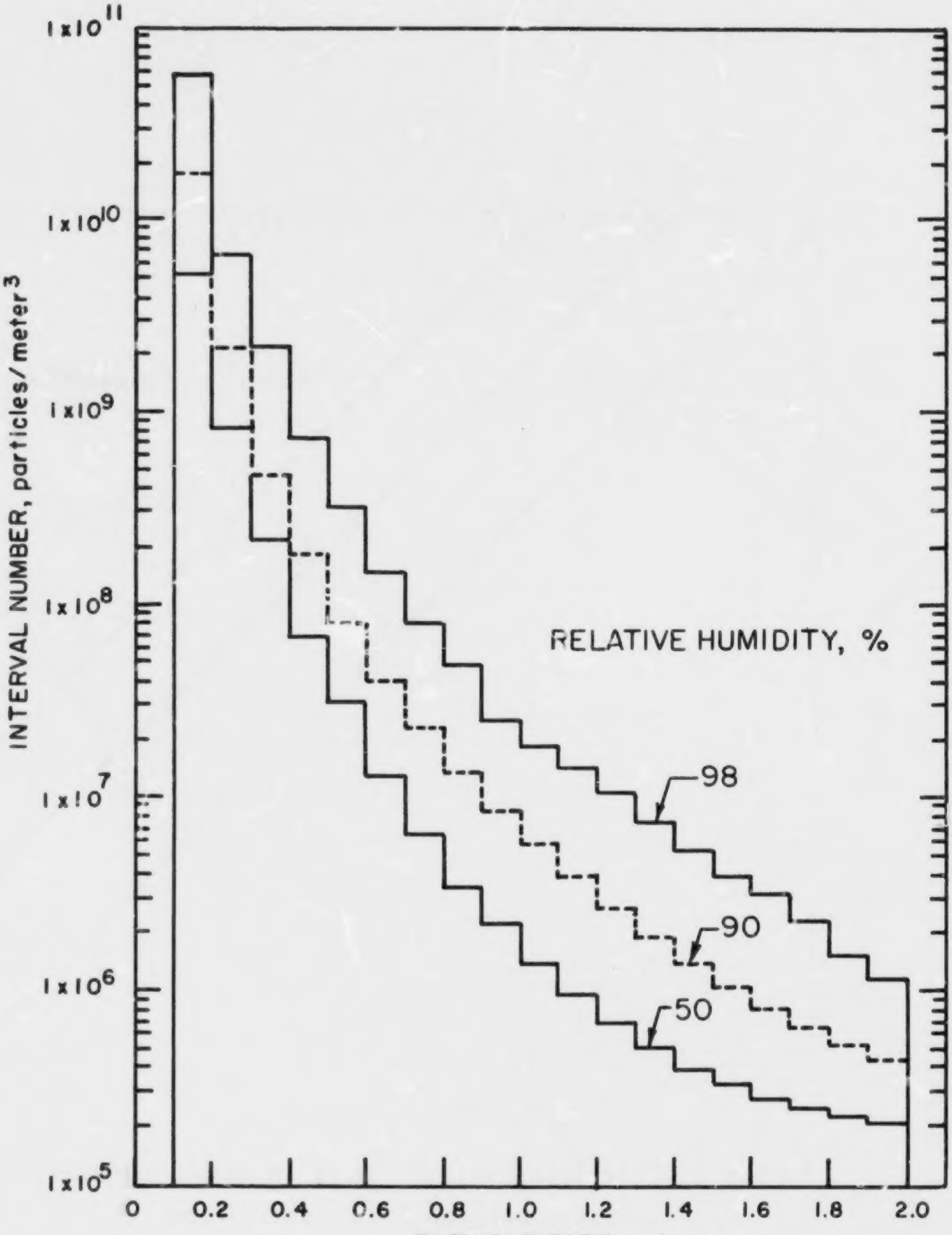

Figure 2. Calculated number of particles per m^3 in different size intervals at different relative humidities in a sample of sulfuric acid mist containing $39 ug/m^3$ of sulfuric acid.

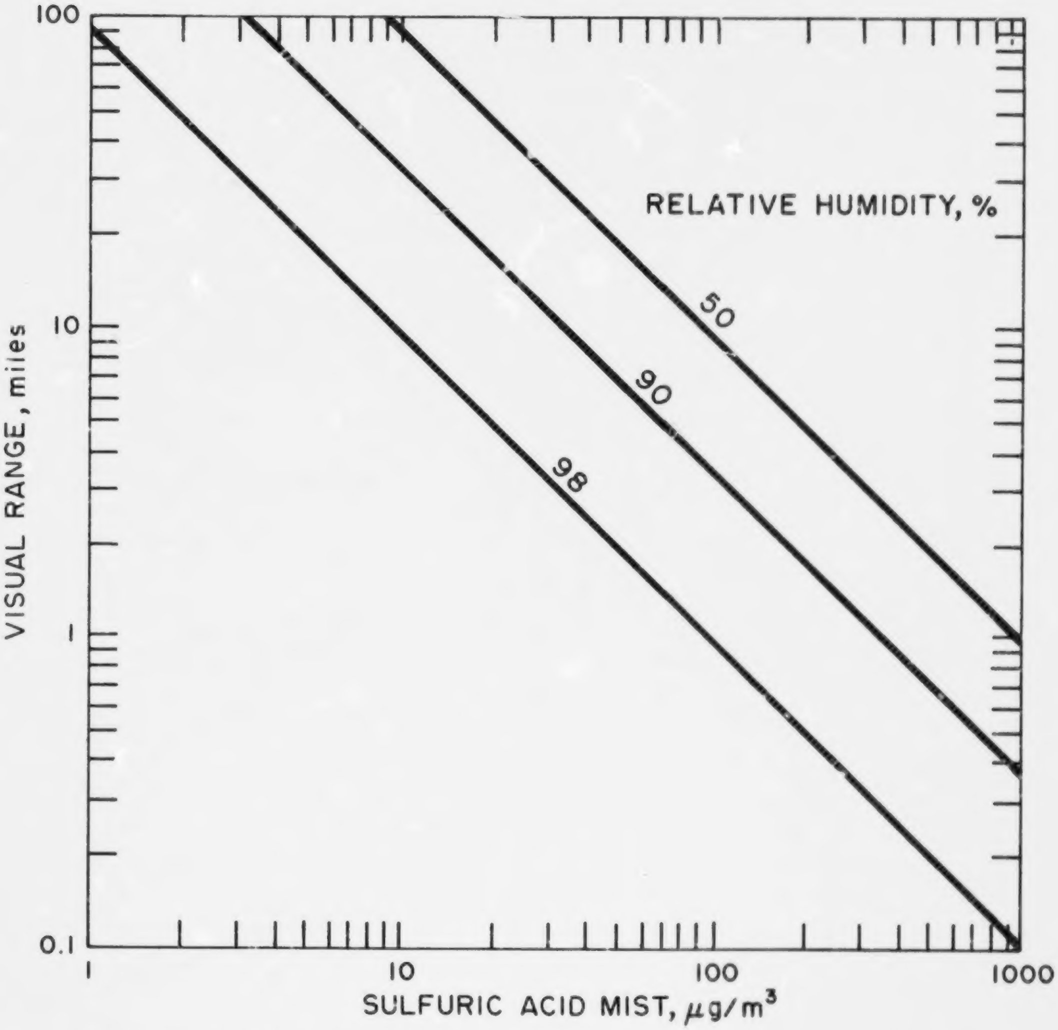

Figure 3. Calculated visual range in miles at various sulfuric acid mist concentrations.

and suspended particulate matter also have been shown, it is possible, given the sulfur dioxide concentration and the relative humidity, to calculate visibility by using the following:

1. Figure 4 shows the concentrations of sulfuric acid mist (measured as H_2SO_4) associated with various concentrations of sulfur dioxide at various relative humidities as calculated from the ratios reported by Bushtueva.[28,29] Ratios similar to those reported by Bushtueva have also been reported by Coste and Courtier,[25,26] Commins,[27] and Thomas.[32]

2. The contribution of sulfuric acid mist to the denominator of the visibility equation $V_2 = \frac{243.6 \times 10^{-5}}{\Sigma N_i A_i E_i}$ varies with relative humidities as follows:

   at 50 percent relative humidity, 0.26×10^{-5} per $\mu g/m^3$;
   at 90 percent relative humidity, 0.69×10^{-5} per $\mu g/m^3$; and
   at 98 percent relative humidity, 2.55×10^{-5} per $\mu g/m^3$.

3. From the National Air Sampling Network data a typical ratio of non-sulfate suspended particulate matter to sulfur dioxide concentration is 1,130 $\mu g/m^3$ to 1 ppm. The components of the suspended particulate matter other than sulfate are assumed non-hygroscopic and their contribution to the denominator of the visibility equation was determined from the investigations of Dennis[113] to be 0.42×10^{-5} per $\mu g/m^3$ of material.

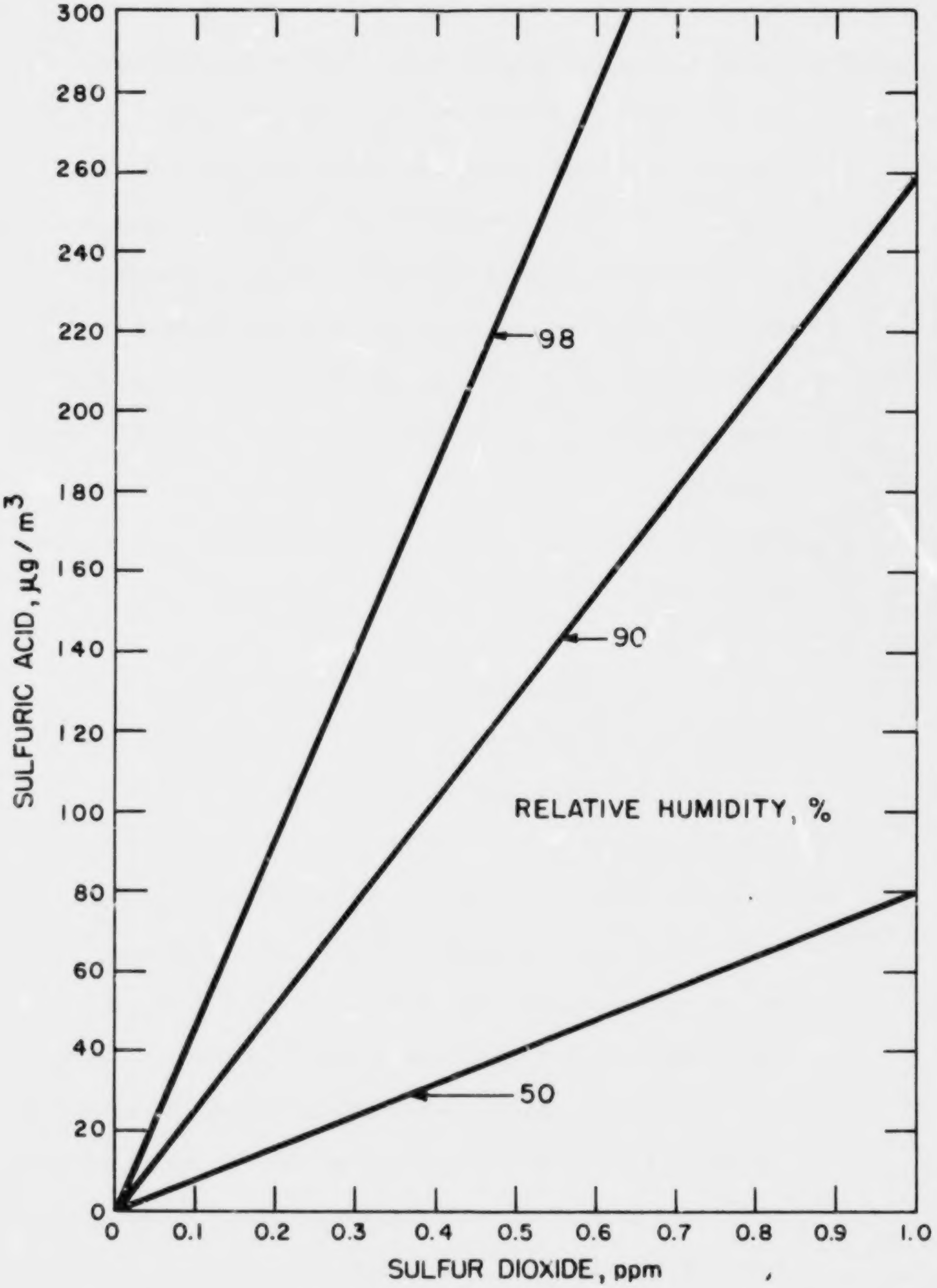

Figure 4. Relationship of sulfuric acid to sulfur dioxide at different relative humidities.

For an example, at 0.3 ppm sulfur dioxide concentration the suspended particulate matter other than sulfate is 0.3 x 1130 or 339 $\mu g/m^3$. At 90 percent relative humidity the sulfuric acid mist content is 78 $\mu g/m^3$.

Then,

$$V_2 = \frac{243.6 \times 10^{-5}}{(339)(0.42 \times 10^{-5}) + (78)(0.69 \times 10^{-5})} =$$

$$\frac{243.6 \times 10^{-5}}{195.22 \times 10^{-5}} = 1.25 \text{ miles.}$$

The results of a series of such calculations are shown in figure 5 which is also reproduced in the summary as figure iii.

A significant effect of reduced light transmission is a reduction in solar energy reaching the ground, which in turn contributes to atmospheric stability and consequently to the further buildup of pollutants. The oxides of sulfur do not in themselves contribute significantly to the loss of solar energy reaching the ground, but other light-attenuating particulates and gases present in the fogs do so. Consequently, fogs last longer, and additional sulfuric acid mist forms, and because of its hygroscopic nature, contributes to longer lasting fogs.[105,114-116]

Acidity of Rain Water and Particulate Matter

Gorham[56] studied the acidity (pH) of rainfall in two cities of England and found that it was more strongly related to the chloride content than to the sulfate content. Such a study apparently has not been made in the United States. However, examination of rainfall samples collected by the National Air Sampling Network shows some pH values around 3.

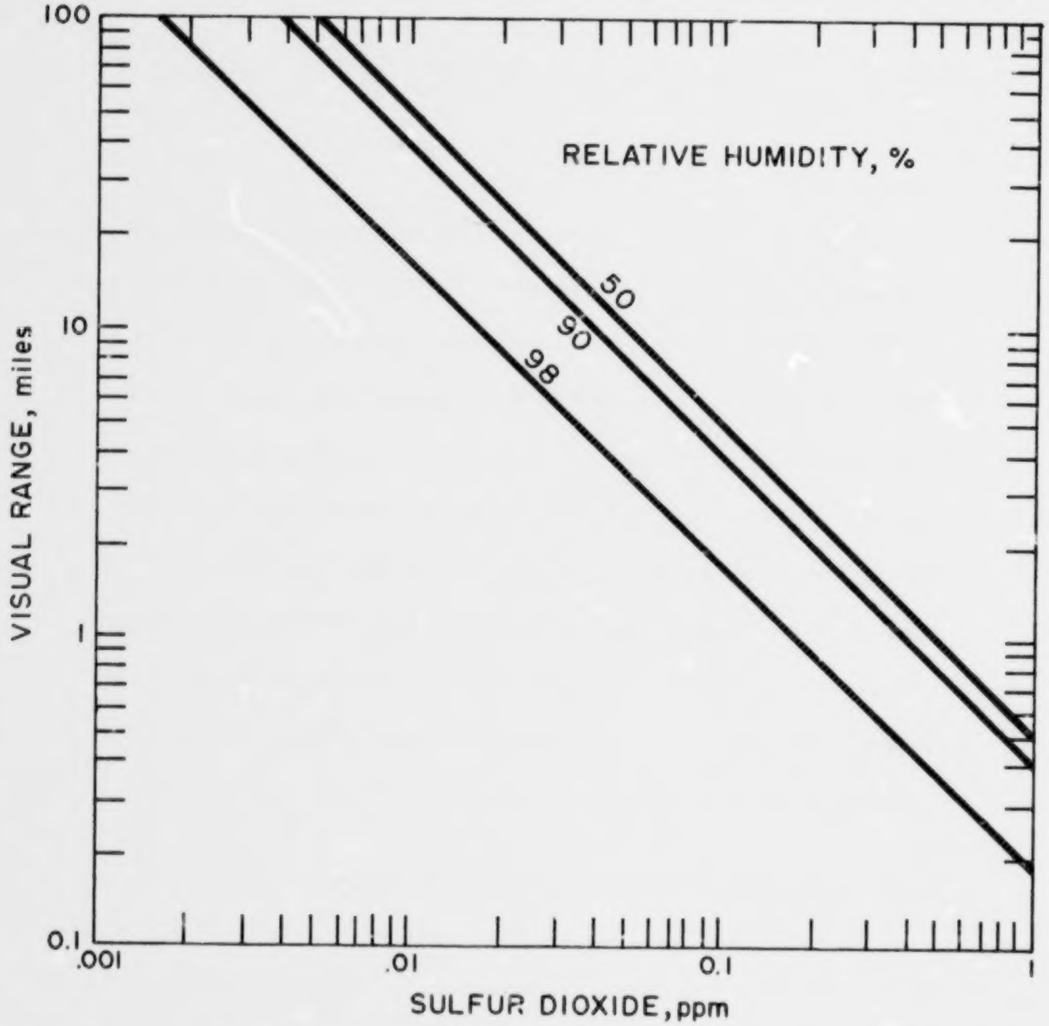

Figure 5. Calculated visual range in miles at various sulfur dioxide concentrations.

Examination of the National Air Sampling Network particulate data indicates that the chloride content of suspended particulate matter is generally very low in atmospheres of the United States.[58] In the one city where there were substantial concentrations of chlorides, unpublished calculations indicate that sulfate and chloride contributed about equally on a normality basis to the pH reading of solutions of particulate matter collected from the air in this city and that an increase of 12 $\mu g/m^3$ of sulfate reduced the pH reading by 0.65. In another set of unpublished data collected by the National Center for Air Pollution Control sulfate was found to account for 21 percent of the variation in pH of solutions of the suspended particulate, with an increase of 12 $\mu g/m^3$ of sulfate reducing the pH by 1 unit. The pH values in both sets of data ranged from approximately 4 to 7.5. In the presence of substantial amounts of ammonia or calcium or other alkaline material, most of the sulfate formed is salt. The similarity of the magnitude of atmospheric sulfuric acid concentrations collected to date, when compared with the magnitude of total sulfate concentrations, indicates that in some atmospheres a large part of the total sulfate may be sulfuric acid.[31-34,70]

Measurements of acidity of dustfall, though seldom reported, indicate that dustfall may be capable of providing a few pounds of hydrogen ion and a few hundred pounds of sulfate ion per acre per year.[75,117-118] The importance of this to soil management is difficult to evaluate because of the heterogeneity of soils, their generally great buffering capacity, and the probable large though unknown buffering capacity of the dustfall itself. An order of magnitude of the effect of acidity in dustfall is that approximately twice as much lime might need to be applied to the soil in areas near large cities with average annual sulfate deposits of 8 to 12 tons per square mile per month (300 to 450 pounds per acre per year) Approximately a tenfold reduction in sulfate deposit would be required to reduce the sulfate deposit to approximately that removed by plants and leaching.[119-120] Even though sulfur in some compounds is a major plant nutrient and is used as a fertilizer element in gypsum under certain conditions, it would seem that uncontrolled deposition of sulfate from air pollution is undesirable.[24]

Effects on Materials

In reviews of the effects of air pollution on materials, Yocom[121] and Burdick and Barkley[123] discuss damage of metals, building materials, leather, paper, and textiles by the oxides of sulfur.

Holbrow[122] found that sulfur dioxide in concentrations from 0.1 to 0.2 ppm can destroy certain paint pigments and, in the presence of ammonia, form crystalline ammonium sulfate bloom. By the author's calculations there is sufficient sulfur dioxide in 2 m^3 of air at 0.1 ppm to give a deposit of 4 $\mu g/cm^2$ over an area of 100 cm^2. They estimate that it would take about 10 m^3 of air to provide the ammonia. Considering that in 1 day lead peroxide candles remove from air containing an average of 0.02 ppm sulfur dioxide the amount contained in 5 m^3, it is reasonable to expect that a moderate bloom could develop in a few days. He found also that concentrations of 1 ppm can delay drying and produce water sensitivity in fresh paint films.

Damage to Metals

Damage to metals by the oxides of sulfur increases with increasing relative humidity and increasing temperature in the normal range of ambient air temperatures. Particulate matter in the air also contributes to the damaging effects. Most, if not all, of the metals corrosion by sulfur oxides is caused by sulfuric acid, either formed from sulfur dioxide in the atmosphere or in droplets on the surface of the metals. The corrosion products are mainly sulfate salts of the exposed metals.[124-127] Gibbons and co-workers[77-79] found among several metals exposed to atmospheres, ranging from rural to heavy industrial, that carbon steels were most affected, followed in descending order by zinc, copper, aluminum, and stainless steel.

Atmospheres polluted with sulfur dioxide and its associated acidity have been found to be among the most corrosive of all atmospheres studied, even more than some marine atmospheres.[128-129] A striking example is the almost fourfold reduction in the corrosion rate of zinc in Pittsburgh associated with approximately a threefold reduction in sulfur dioxide, 0.15 to 0.05 ppm, and a twofold reduction in dustfall in the period 1926 to 1960.[101,128]

Gibbons and co-workers[77-79] found rather direct relationships between corrosion and oxides of sulfur pollution in several ambient atmospheres in which annual average lead peroxide candle sulfation rates ranged from approximately 0 to 12 mg SO_3/100 cm^2/day. The increase in corrosion per unit increase in sulfation was greater at lower sulfation rates; e.g., of the maximum corrosion noted at 12 mg SO_3/100 cm^2/day approximately one-half had occurred with iron and about one-fifth with zinc at 2 mg SO_3/100 cm^2/day. In terms of the measurements made by these investigators, a lead peroxide sulfation rate of 2 mg SO_3/100 cm^2/day represents an average of 0.056 ppm of sulfur dioxide. Thus, at annual average sulfur dioxide concentrations even lower than 0.05 ppm, considerable corrosion takes place, particularly with iron.

The practical significance of metals corrosion has not been given adequate attention. It has been estimated that in England about one-third of the annual replacement cost for steel rails is caused by air pollution.[130] Couy[131-133] investigated the effect of atmospheric corrosion on maintenance and economics of overhead line hardware and guy wire strand in Pittsburgh. The conditions of actual materials in use over the period roughly 1920 to 1940 were determined for areas of severe and average pollution. Areas of severe pollution included valleys traversed by railways and areas in direct wind-line with industrial fumes. The author developed formulas to determine the amount of inspection required, the materials needed, and the costs involved for a particular situation. No direct cost estimates were made but it appears that life of the materials in the severely polluted areas was reduced to about two-thirds of that of the materials in the average polluted areas. Average annual sulfur dioxide pollution during the period of the study was initially very high (0.15 ppm) but had declined to approximately 0.05 ppm by 1940 (0.04 ppm in 1963).[101] Gilbert[134] reported that the lives of zinc and aluminum facings in industrial Pittsburgh during the period of the study were 7 and 10 years, respectively, compared to an expected 30 to 40 years for each in a rural atmosphere.

The effects of air pollution on electric contacts result in increased costs because of the losses associated with increased resistance of filmed contacts and because more costly, less reactive metals must be used. The use of gold instead of silver, for instance, costs about 14.8 million dollars annually. If gold could be replaced by palladium, a saving of 8 million dollars would result, but palladium tarnishes in a sulfur dioxide atmosphere.[135]

Effects on Other Materials

There is no doubt that air polluted with oxides of sulfur is detrimental to portland cement, various stones, paper, leather, and textiles. However, there has been little recent investigation of these effects, and early work dealt only with high concentrations of sulfur dioxide. The magnitude or practical significance of sulfur oxides pollution on these materials needs to be investigated.[25,123,136-137]

Salvin[138-139] studied color changes of dyed fabrics exposed without sunlight to the atmospheres of Los Angeles, California; Chicago, Illinois; Phoeniz, Arizona; and Sarasota, Florida during the period October through December 1961. The atmosphere which caused the most fading was that of Chicago where sulfur dioxide concentrations were highest (NASN annual average about 0.09 ppm).[33-34]

Effects of Sulfur Dioxide on Vegetation

Recent and substantial reviews of injury to vegetation by the oxides of sulfur have been made by Brandt,[140] Thomas,[141] and Scheffer and Hedgcock.[100]

According to Thomas, the effects of sulfur dioxide on plants are fairly well understood, with two types of injury, from acute and chronic exposure, being recognized.

The gas is absorbed into the mesophyll of the leaves through the stomata. Toxicity is due largely to the reducing properties of the gas. The limiting concentration that can be tolerated in the cells is about the same for many diverse species, including water plants. When this concentration is exceeded, the cells are first inactivated with or without plasmolysis, then killed. When extensive areas are killed, the tissues collapse and dry, leaving a characteristic pattern of interveinal and marginal acute injury.

If only a few cells in an area of a leaf are injured, the area may become chlorotic or brownish red in color, owing to chronic injury. Chronic injury is characterized by a slow oxidation of sulfite to sulfate in the cells. Consequently, if sulfur dioxide is not added rapidly to the system, a rather large total amount may be added before sulfate toxicity occurs. Sulfate toxicity is a form of chronic injury manifested by white or brownish-red turgid areas on the leaf caused by rupture of some cells. Abscission of leaves often occurs at or before this stage.

Few studies indicate that plants are injured when exposed to sulfur dioxide concentrations lower than those required to produce visible leaf injury. For example, in an exposure of alfalfa to 0.10 ppm for 45 days, there was no significant effect on growth or chlorophyll content, although there was some abscission of older leaves. New growth maintained the activity of the plants.[141] However, Bleasdale[142] found that the growth of a rye grass Lolium perenne (Aberystwyth S23), in Manchester, England, was definitely retarded in the open atmosphere or in a greenhouse supplied with ambient air compared with growth in a greenhouse supplied with air scrubbed by water sprays. Ambient sulfur dioxide concentrations were 0.05 to 0.20 ppm. No evidence of acute leaf injury was seen in the greenhouse with unscrubbed air, but the rate of senescence of the plants was greater than in washed air, and rate of tiller formation, number of leaves, and dry weight were reduced by the pollution.

Generally, the plants most sensitive to sulfur dioxide are those with succulent leaves having high physiological activity, plants such as alfalfa, the grains, squash, cotton, grapes, and endive. Exposure to approximately 1.25 ppm sulfur dioxide for 1 hour causes incipient injury to alfalfa, barley, endive, and cotton.

Leaves with a heavy waxy or cutinized epidermis, such as citrus, pine, and privet, are generally more resistant.

High light intensity (especially in the morning hours), high relative humidity, adequate moisture supply, and moderate temperatures predispose plants to injury because these factors cause the stomata to open, thereby facilitating absorption of sulfur dioxide.

Under conditions of maximum susceptibility, injury to alfalfa and equally sensitive plants can begin with exposures to sulfur dioxide at 0.28 ppm averaged over 24 hours. The following equations, in which "C" is the concentration of sulfur dioxide in ppm and "t" is the time of exposure in hours, apply to alfalfa for single exposures to relatively high concentrations:

if $(C-0.24)t = 0.94$, traces of leaf destruction appear.

if $(C-1.4)t = 2.1$, 50 percent leaf destruction occurs.

if $(C-2.6)t = 3.2$, 100 percent leaf destruction occurs.

Thus, injury begins after 4 hours of exposure to 0.48 ppm, etc. With alfalfa the reduction in yield is proportional to the amount of leaf tissue destroyed.

Scheffer and Hedgcock reported injury to some species of trees and shrubs during an exposure to 0.5 ppm of sulfur dioxide for 7 hours. They concluded that the lowest concentration of sulfur dioxide that injures conifers is not less than 0.25 ppm.

In the past the greatest concern for injury to plants by sulfur oxides pollution has been concern with large sources, such as smelters. Scheffer and Hedgcock reported that in 1910 and 1911, all of the major tree species were either dead or dying at

5 to 8 miles from the smelter in Anaconda, Montana. Outside a 22-mile radium from the smelter, no injury was observed.

In the vicinity of the Trail, British Columbia, smelter, which in 1929 emitted an average of 9,300 tons of sulfur (18,600 tons of sulfur dioxide) per month, plant injury was noted as far as 52 miles south of the smelter. Three zones of injury were delineated on the basis of the percent injury to ponderosa pine, Douglas fir, and forest shrubs: Zone 1 equalled 60 to 100 percent injury; Zone 2 equalled 30 to 60 percent injury; and Zone 3 equalled 1 to 30 percent injury. Zone 1 in which injury was acute extended about 30 miles south of the smelter in a river valley; Zone 3, at higher elevations, extended 52 miles south of the smelter and contained trees with relatively slight markings and trees suffering from slow but progressive deterioration. After control devices were installed, measurements in 1934 and again in 1935 showed that the injury to broadleaf trees and to shrubs had dropped to 20 percent in Zone 1 and to 4 percent in Zone 3. Appraisal of ponderosa pine cone production in 1936 revealed that 81 percent of the pines in Zone 1 had no cones and that outside the zone 16 percent of the pines had no cones. Sulfur dioxide concentrations 15 miles south of the smelter in Zone 1 averaged about 0.03 ppm during the summer with occasional peak concentrations in excess of 0.5 ppm for a total time of 5 to 10 hours.

Linzon[83] reported marked effects on growth and needle color of white pine at locations 25 miles and less from the Sudbury, Ontario, smelter where measurable sulfur dioxide concentrations were present from 10 to 20 percent of the time. Concentrations above 0.25 ppm of sulfur dioxide were present from less than 1 to 4 percent of the time; the remainder of the measurable concentrations were less than 0.25 ppm. The annual average concentrations apparently ranged downward from 0.03 ppm.

Sullivan[81] reported damage to 36 percent of the home gardens in an area near a smelter where the annual average sulfur dioxide concentration was 0.009 ppm and the maximum daily average 0.155 ppm. Eighty-nine percent of the gardens were damaged in another area more influenced by the smelter; there the annual average was 0.033 ppm and the maximum daily average 0.60 ppm. In the latter area, during brief fumigations sulfur dioxide concentrations ranged up to 5 to 10 ppm.

Linzon[82] reported injury to trees in the vicinity of petroleum refineries refining crude oil containing from 0.5 to 2.5 percent sulfur. Measured concentrations of sulfur dioxide in the atmosphere exceeded 0.5 ppm during 10 hours in 1 month with momentary peaks over 2 ppm.

Injury can occur where annual average concentrations are very low when the sources of sulfur dioxide and/or the meteorologic conditions are such that exposures in excess of the threshold occur. There is some evidence of injury, particularly chronic injury, where concentrations never exceed 0.20 ppm, but in general exposures of 8 or more hours duration to concentrations in the range of 0.20 to 0.30 ppm of sulfur dioxide generally appear to be necessary for incipient injury to vegetation.

At the present time injury to plants from many widely distributed sources of sulfur dioxide are of concern. Markings of sulfur dioxide injury may be found on plants within most industrial communities and around most coal or residual oil-burning power-generating plants. Although sulfur dioxide by itself may not be responsible, a large amount of the white pine in eastern Tennessee has been lost or is dying from exposure to emissions from coal-fired power plants.[143]

Effects of Sulfuric Acid Mist on Vegetation

Thomas et al.[144] discussed experiments in which plants were exposed to sulfuric acid mists in concentrations of from 108 to 2,160 mg/m^3. Sulfuric acid droplets can settle on dry leaves without causing injury, but when the leaf surface is wet a spotted injury develops.

Middleton et al.[145-146] and Thomas[144] reported that this type of injury occurred in the Los Angeles area during periods of heavy air pollution accompanied by fog when the surface of the leaf may be wet. This type of injury may also occur in the absence of fog near combustion effluents containing sulfur oxides when the gas effluent dew point permits acid droplet formation.

The sequence of symptom development is one in which the exposed surface, usually the upper surface, shows the initial necrosis. The pH of the leaf-surface moisture may be less than 3. Cellular collapse in many small spots develops progressively through the upper epidermis, mesophyll, and lower epidermis of the leaf, leaving scorched areas. No glazing or bleaching accompanies this injury, and leaf areas covered by exposed leaves show no markings. In the Los Angeles area swiss chard and beets were reported to be most typically injured. Alfalfa also develops a spotted injury pattern. Spinach is more uniformly wet by fog and shows a more diffuse type of injury.

EFFECTS OF OXIDES OF SULFUR ON MAN AND ANIMALS

Evidence for the effects of air pollution on animal and human health comes from several sources: known toxicological properties of compounds and mechanisms of action; physiological responses and pathological developments observed during exposures of experimental animals to pollutants; industrial exposures of humans; human exposures to experimental atmospheres; clinical changes in individuals exposed to changes in ambient air pollution; epidemiological studies of mortality and morbidity in relation to air pollution; studies of air pollution disasters. General reviews of the subject have been made by Heimann,[147] Goldsmith,[148] Lawther,[149] Phair,[150] Phillips,[151] Catcott,[152] and Stokinger.[153] In addition, a number of shorter papers report the health effects of polluted air which contained oxides of sulfur along with other pollutants.[154-172]

Acute Toxicity from Excessively High Exposures

Brief general descriptions of the acute toxicity of sulfur dioxide at high concentrations (100 to several thousand ppm) are available.[173-176] The effects of inhalation of oxides of sulfur by man and animals were reviewed by Greenwald,[177] Amdur,[5] and Setterstrom.[178] The acute toxicity of sulfur dioxide to rats and mice was studied in detail by Weedon et al.[179] and the acute toxicity of sulfuric acid mist was studied by Treon et al.[180] and by Pattle et al.[181] Around 1,000 ppm of sulfur dioxide in air are required to kill mice, rats, rabbits, and guinea pigs during 8 to 16 hours of exposure. Several hundred milligrams of sulfuric acid mist per cubic meter are required to kill mice, rabbits, and rats, but as little as 18 mg/m^3 has killed guinea pigs. Sulfuric acid mists of 2.7μ MMD were more toxic than those of 0.8μ and mists of a given particle size were more toxic at 0°C than at room temperature.[181]

McCallan and Setterstrom[182] found on the basis of the time required for 1,000 ppm of sulfur dioxide to kill 50 percent of exposed organisms consisting of various fungi, bacteria, plants, seeds, insects, and rodents that plant leaves were most sensitive to the gas followed by fungi and bacteria, plant stems, animals, plant seeds, and sclerotia of fungi, in that order.

Basic Studies with Sulfur Dioxide

Several basic studies of sulfur dioxide have been pursued.

Thompson and Pace[183] exposed various tissue culture cell lines to sulfur dioxide at 5 and 25 ppm for 8 hours a day for 2 days and found that at 25 ppm all cells were killed when directly exposed, whereas at 5 ppm only detached single cells were killed.

Dalhamm and co-workers[184-189] studied the effect of sulfur dioxide on ciliary activity and mucous flow in isolated tracheae and through windows which they made in the tracheae of live animals. Ciliary movement in isolated trachea ceased after 25 minutes of exposure to 7 ppm; but in single exposures of the tracheal epithelium in live animals to 100 ppm or less of sulfur dioxide ciliary activity usually was not affected, although in a few animals it was decreased slightly. Carbon particles mostly below 1µ in size and in concentrations of 1.6 to 2.3 mg/m^3 did not affect ciliary movement. Simultaneous inhalation of carbon particles with 100 ppm sulfur dioxide resulted in decreases in ciliary activity similar to those with sulfur dioxide alone. After exposures of live animals for 18 or 67 days to approximately 10 ppm of sulfur dioxide, ciliary activity was diminished in some animals, and in all animals flow of mucus was greatly reduced, the mucous blanket was thickened and tenacious, and epithelial crypts were demonstrated in the tracheae.

Reid[190] found no pathological change in the lungs of rats exposed to 40 ppm of sulfur dioxide for 5 hours a day, 5 days a week. However, a number of rats exposed to 300 to 400 ppm for varying periods up to 3 months showed an increased amount of mucus by the fourth week. In the latter weeks of the 3 month exposure, positive bacterial cultures could be isolated only from the trachea, bronchi, and lungs of exposed rats. After 2-1/2 weeks of exposure, an increase in the number of goblet cells in the respiratory tract was detected. Three months after cessation of exposure the increased number of cells persisted, but a decline in the secretion of mucus was observed.

Balchum and co-workers,[191-192] Dalhamn and co-workers,[184-189] and Strandberg[193] studied the absorption, retention, and distribution of sulfur dioxide in the respiratory tract of animals by various methods. At 700 ppm only 5 percent of the sulfur dioxide reaches the bronchi, whereas at 0.05 ppm 60 percent of the sulfur dioxide has been shown to reach the bronchi. According to Strandberg,[193] the solubility of sulfur dioxide decreases with decreasing concentrations. Differences in solubility at various concentrations may account for the differences at various concentrations in the percentage of sulfur dioxide reaching the bronchi. In exposures of humans to 16 ppm sulfur dioxide for 30 to 45 minutes, Frank and Speizer[194] determined that less than 2 percent of the gas entering the nose reached the pharynx. However, as a result of desorption from the mucosal surface, final net uptake was 84 percent of the dosage. Leong and MacFarland[195] reported 27.2 percent uptake in rats exposed to 750 ppm sulfur dioxide for 2 hours and 51.8 percent uptake after 2 hours exposure to 41 ppm. The percent uptake decreased with time of exposure and increased as concentrations decreased. Studies with radioactive sulfur dioxide ($S^{35}O_2$) show that sulfur is distributed to all parts of the body and then slowly eliminated through the lungs, kidneys, and intestines.[191-192, 196-199] Continued inhalation results in body accumulation of labeled sulfur.[199]

Nadel et al.[200] administered an unmeasured amount of sulfur dioxide through tracheal cannulae to the upper and lower airways of anesthetized cats. Sulfur dioxide delivered into the lower airways and lungs of 11 cats during a single inflation increased airway resistance by an average of over 200 percent. Sulfur dioxide delivered to the upper airways (upper cervical trachea, larynx, and oropharynx) of 7 cats increased airway resistance by an average of over 300 percent. Sulfur dioxide had no demonstrable effect on the lower or upper airways when administered during cooling of the vagosympathetic nerves or when administered after intravenous injection of atropine sulfate. Thus, sulfur dioxide appears to produce these observed effects via nerve pathways which stimulate receptors to produce reflex bronchoconstriction.

Exposures to approximately 150 ppm of sulfur dioxide for 3 hours have resulted in marked drops in vitamin C content of the blood of rabbits.[201] Similar exposure of guinea pigs for 3 to 6 days resulted in the development of acute gastric ulcers.[202]

Effects on Lung Function of Brief Exposures of Animals to Sulfur Oxides

Salem and Aviado[203] exposed anesthetized dogs to 200 to 850 ppm of sulfur dioxide and noted pulmonary vasoconstriction, bronchoconstriction preceded by bronchodilation, increased pulmonary arterial blood pressure, depression of myocardial force of contraction accompanied by bradycardia, and systemic shock. Balchum and co-workers[191-192,196-197] exposed anesthetized dogs to sulfur dioxide in concentrations from 1 to 150 ppm for 20 to 40 minutes. The animals breathed the gas either through the nose and mouth or through tracheal openings. In both situations there were decreases in lung compliance and increases in respiratory resistance with all concentrations of sulfur dioxide. The increase in nonelastic resistance occurred within 9 seconds after the onset of breathing sulfur dioxide and disappeared quite as quickly following the end of exposure. However the adverse effects were greater in animals breathing through the tracheal opening. Amdur and co-workers[204,205] observed increased airway resistance in guinea pigs during exposures for 1 hour to sulfur dioxide in concentrations from 2 ppm to approximately 1,000 ppm. The increase in resistance ranged from 20 percent at 2 ppm to 300 percent at about 1,000 ppm.

Amdur and co-workers[204-211] continued their pulmonary function studies by exposing guinea pigs to : (1) sulfuric acid mists; (2) combinations of 82 ppm sulfur dioxide with 19 mg/m^3 of 0.8μ-size sulfuric acid mist and 120 ppm sulfur dioxide with 8 mg/m^3 of 2.5μ-size sulfuric acid mist; (3) sulfate salts from 0.25 to 3.6 mg/m^3 of various particle sizes from 0.29 to 1.4μ; (4) the combination of 2 ppm sulfur dioxide with 0.25 mg/m^3 of 0.29μ-size zinc ammonium sulfate; and (5) 0.5 to 1,000 ppm sulfur dioxide with 4 to 10 mg/m^3 of sodium chloride of 0.04 and 2.5μ-size.

It was concluded from these experiments that: (1) simultaneously inhaled particulate matter, such as sodium chloride which itself has no demonstrable effect on airway resistance, does enhance the effect of sulfur dioxide; (2) compared with sulfuric acid mist or sulfate salts (zinc sulfate, ammonium sulfate and zinc-ammonium sulfate) on the basis of weight per volume of air, 20 to 200 times as much sulfur dioxide is required to produce a 50 percent increase in airway resistance; (3) in the size range 0.04 to 7µ the smaller particles increase airway resistance more or enhance the effect of sulfur dioxide more than the larger particles; (4) the increase in airway resistance resulting from exposure to sulfur dioxide in combination with irritant particles is greater than the the sum of the responses to each agent alone; and (5) increased airway resistance resulting from exposure to irritant particles or to sulfur dioxide in combination with particles lasts for much longer periods after discontinuation of exposure than the increased airway resistance with sulfur dioxide exposure alone.

Effects of Prolonged Exposures of Animals to Oxides of Sulfur

Information on prolonged exposure of animals to oxides of sulfur is relatively scarce. Weedon et al.[212] exposed mice and guinea pigs to concentrations ranging from 10 to 35 ppm of sulfur dioxide for up to 47 days (concentrations ranging from 60 to 1,000 ppm which were also used, killed some animals). They concluded that no significant mortality or signs of distress occurred among healthy animals at concentrations of 33 ppm or below. Heyssel and co-workers[213-214] exposed rats over most of their lifetime to concentrations of 1, 2, 4, 8, 16, and 32 ppm of sulfur dioxide. Their data indicate that over this range of concentrations life during exposure was significantly reduced by 0.08 months for each 1 ppm increase of sulfur dioxide concentration. The authors also observed wheezing, development of eye opacities, loss of fur, appearance of scaley tails, and elevation of hemoglobin concentrations and white blood cell counts, especially the neutrophils, when animals were exposed to increased concentrations of sulfur dioxide.

Navrotskii[215] exposed 20 rabbits to 7 ppm of sulfur dioxide 2 hours per day over 5.5 to 8.5 months. Rabbits which were immunized against typhoid either after or during exposure showed a substantial drop in agglutination titers. No significant change was demonstrable in blood complement or in blood protein fractions.

Prokhorov and Rogov[216] exposed 11 rabbits to 70 ppm of sulfur dioxide for 3 hours per day for 13 weeks and noted changes in the parenchyma of various organs and increased permeability of vascular walls. The authors further reported decreased activity of free sulfhydryl groups in the brain and, when carbon monoxide (160 to 320 ppm) was added to the exposure atmosphere decreased succinic dehydrogenase activity resulted.

Lobova[217] exposed 10 rats to 0.48 mg/m^3 (0.16 ppm) of sulfur dioxide 4 hours per day for 114 days. The exposure did not affect body weight but dehydrogenase activity of the spleetn was reduced by 50 percent and there was a lesser but general reduction in dehydrogenase activity of muscle, liver, lung and heart tissues. Cholinesterase activity was reduced in the spleen, only slightly in other organs and was not affected in the peripheral blood. Vitamin C content of the small intestinal mucosa, and of the liver and kidneys was reduced. Rats exposed to 0.1 mg/m^3 (0.03 ppm) sulfur dioxide 5 hours per day for 166 days showed reduced blood carbonic anhydrase activity between the 141st and 162nd day of exposure, but the effect was reversible upon discontinuation of exposure.

Bushtueva[218-219] reported that guinea pigs exposed to 2 mg/m^3 of sulfuric acid mist for 5 days (no particle size given) developed circulatory and lymphatic distrubances as evidenced by focal edema. Other pathological changes noted were edema of lungs and intra-alveolar wall thickening. After 1 to 2 weeks exposure to approximately 1 mg/m^3 of sulfuric acid, a slight catarrhal condition was observed in the mucous membranes of the trachea and bronchi. Widely distributed, but only slightly defined, interstitial proliferative processes accompanied by round lymphoid cell infiltration surrounding the blood vessels and bronchi also appeared. This latter pathological process was more highly developed when animals were observed 2 to 3 months after discontinuation of the exposure. With 0.5 mg/m^3 of sulfuric acid lung irritation was slight. But when the exposure consisted of 0.5 mg/m^3 of sulfuric acid mist plus 0.3 ppm of sulfur dioxide, thickening of the interalveolar septi, formation of lymphatic folliculi, signs of collagen formation in the form of perivascular and peribronchial fibrosis, and fibrosis in the interalveolar septi appeared 2 months after discontinuation of the exposure. Histamine content of the lungs increased parallel with the pathology. Similar results were obtained by Thomas et al.[220] when they exposed guinea pigs to up to 4 mg/m^3 of sulfuric acid mist of 0.6, 0.9, and 4μ diameter for 18 to 140 days; the 0.9μ sized particle had the greatest effect.

Prolonged Exposures of Animals to Mixtures Containing Oxides of Sulfur

In addition to the previously mentioned experiments in which the exposure atmospheres contained more than one agent, Bushtueva[221] chronically exposed rats to low concentrations of sulfur dioxide, sulfuric acid, and combinations of the two. She concluded that the results are additive.

Cameron[222] exposed goats, rabbits, guinea pigs, mice and monkeys to two chlorosulfonic acid/sulfur trioxide mixtures, one containing approximately 30 mg/m^3 of sulfur trioxide and 1.8 mg/m^3 of hydrochloric acid and the other containing approximately 60 mg/m^3 of sulfur trioxide and 3 mg/m^3 of hydrochloric acid. Animals were exposed to the first mixture for 6 hours each day for 14 days and animals were exposed to the second mixture for 6 hours each day for 9 days. Some guinea pigs were killed by the mixture containing the lower concentrations after only 6 hours.

The above data indicate the types of patho-physiological responses which may result from brief exposure of laboratory animals to one or two of the oxides of sulfur and other known substances at high concentrations. However, the data do not necessarily indicate the type of adverse effects that may result from exposure to ambient air containing various other types of pollutants.

Drinker[223] suggested that the effects from experimental exposures to coal smoke would more closely approximate the effects of ambient air polluted with oxides of sulfur. Few such experiments have been reported in the literature. Schnurer[224] exposed four rabbits and eight rats 23 hours per day for 80 days and four mice for 17 days to smoke from anthracite (0.48 percent sulfur), coke (0.92 percent sulfur) and bituminous coal (1.36 to 1.53 percent sulfur). The exposure atmosphere produced by anthracite smoke contained 1.91 ppm sulfur dioxide and 3.12×10^8 particles/m^3; the exposure atmosphere produced by coke smoke contained 9.12 ppm sulfur dioxide and 3.7×10^8 particles/m^3; and the exposure atmosphere produced by bituminous coal contained 7.51 ppm sulfur dioxide and 4.41×10^9 particles/m^3. For the particle counts, samples were taken with an Owens jet counter, and the counts were made under a microscope at a magnification of 970. The hemotological studies showed increases in the red and white blood cell counts and in the percentage of hemoglobin. The greatest increase was noted in animals exposed to bituminous coal smoke and the smallest increase in animals exposed to anthracite coal smoke. Some animals from each group were killed 2-1/2 months after, and others were killed 14 months after the exposures were terminated. No significant pathology could be detected in the lungs of animals exposed to coke or anthracite coal smoke, but animals exposed to bituminous coal smoke developed evidence of fibrosis, proliferation of the bronchial epithelium, and marked peribronchial lymphoid hyperplasia.

Vintinner and Baetjer[225] exposed rats to bituminous coal dust and to filtered and unfiltered bituminous coal smoke for various periods of time up to 165 days. Sulfur dioxide concentrations in the coal smoke ranged between 0.7 and 1.6 ppm. Total particulate matter ranged from 10.6 to 95.5 mg/m^3 in the filtered smoke and from 8.2 to 262 mg/m^3 in the unfiltered smoke. Either 7 days before the end of exposure or on the last day of exposure the animals were intrabronchially inoculated with a human strain of Pneumococcus Type 1. There were no significant differences between the total incidences of infection nor in the case fatality rates of smoke exposed and control animals. However, the strain of rats used in the study normally develop areas in the lungs which are filled with mucus, pus or caseous material and the incidence of this condition was 40 percent in rats exposed to smoke for more than 20 weeks as compared to 20 percent in the controls.

Pattle and Burgess[226] exposed mice and guinea pigs to sulfur dioxide concentrations of approximately 1,000 ppm, to 50 to 100

mg/m^3 of smoke derived from a kerosene lamp, and to combinations of both. All animals exposed for 66 hours to approximately 50 mg/m^3 of either kerosene smoke or kerosene smoke with "absorbed" sulfur dioxide survived the exposures, and lung examination revealed no signs of edema, consolidation, hemorrhage or emphysema in either group; however, there was slight capillary congestion in both groups. When mice had previously been exposed to smoke, less edema was produced in their lungs when they were exposed to sulfur dioxide than otherwise. Exposure of mice simultaneously to sulfur dioxide and smoke resulted in quicker deaths than exposure to sulfur dioxide alone; this was not true with guinea pigs. Salem and Collumbine[227] and Pattle and Collumbine[228] performed similar experiments but included sulfuric acid mists of 1.2 and 0.5μ in concentrations of 50 to 150 $\mu g/m^3$. They reported that the toxicity of sulfuric acid was increased by the simultaneous addition of smoke and that, in contrast to sulfur dioxide, exposure to smoke prior to exposure to sulfuric acid did not alter the toxicity of sulfuric acid.

Industrial Exposures of Humans to Sulfur Oxides

Accidental industrial exposure to fuming sulfuric acid was reported to have resulted in immediate respiratory difficulty which eventually led to disabling pulmonary fibrosis, residual bronchiectasis, and pulmonary emphysema.[229]

Studies dealing with occupational exposure to sulfur dioxide in varying concentrations of 10 to 50 ppm produced variable findings.[230-235]

Anderson[230] studied oil refinery workers in Iran where the temperature is high and the humidity low. The workers were exposed primarily to sulfur dioxide in concentrations that ranged usually from 0 to 25 ppm but which peaked as high as 100 ppm. Anderson could not demonstrate adverse effects in these employees; however, the studies included only measurement of vital capacity, routine chest x-rays, body weight, and blood pressure.

Kehoe et al.[231] in 1932 studied 100 workmen engaged in handling sulfur dioxide in refrigeration plants. Sulfur dioxide concentrations were usually around 25 ppm with occasional concentrations averaging 80 to 100 ppm. X-rays showed nothing to indicate the existence of injury to the lungs or bronchi. However, the investigators observed symptoms of irritation of the upper respiratory tract as manifested by nose bleeds, coughs, hemoptysis, and constriction of the chest. Symptoms associated with severe exposure were those of coughing, sneezing, eye irritation, sore throat, chest pain, and loss of appetite. Other significant observations noted were higher incidence of abnormal acidity of urine, tendency to increased fatigue, shortness of breath on exertion (dyspnea), abnormal reflexes and increased duration of colds.

Evans[232] studied x-rays of lungs of 18 employees whose average age was 44 years and who were exposed to sulfur dioxide and sulfur trioxide in the manufacture of sulfuric acid for an average of 10.7 years. All cases had x-ray evidence of fibrosis, but there was no demonstrable roentgenological evidence of progression of the disease over the previous 4 to 5 years.

Skalpe,[233] studied Norwegian pulp mill workers in Norway who were exposed to 2 to 36 ppm sulfur dioxide for from 1 month to 45 years. He found a significantly increased frequency of cough, expectoration, and dyspnea on exertion. The difference in frequency was greater in the group under 50 years of age. In this age group the maximum expiratory flow rate was significantly lower than in the control group. Vital capacities were not significantly different between exposed and control groups.

Viikeri[234] in Germany studied x-ray changes in lungs of 53 foundry workers who had been exposed for more than 10 years to an average of less than 10 ppm of sulfur dioxide, but with occasional higher exposures. The x-rays showed normal lungs in only 7 of 53 exposed workers, whereas in the control group 20 out of 37 were normal. The chief pathological findings in the 53 exposed workers were 4 cases of fibrosis, 17 cases with enlarged hilum, and several cases of emphysema and cardiac disease. Approximately one-third of both the exposed and control groups had evidence of old pulmonary tuberculosis.

Litkens[235] studied workers in metallurgical plants who were exposed to 0.015 to 1.5 mg/l of sulfur dioxide (5 to 500 ppm). The author concluded that both increasing concentrations of sulfur dioxide in the working atmosphere and increasing durations of exposure resulted in increasing laryngitis, pharyngitis, chronic bronchitis, pulmonary fibrosis, and emphysema. The author also concluded that interference with enzyme systems by inhaled sulfur dioxide circulating in the blood resulted in the pathology. There were also increased incidences of pneumonia and gastrointestinal disorders.

Other reported responses to sulfur dioxide in industrial exposures are allergic skin reactions[236] and asthma.[237-238]

In these reported industrial exposures no observed effects were noted in some cases whereas in other cases there was evidence of deep lung damage, including fibrosis and emphysema. The studies indicate that less-serious effects are produced by low humidity atmospheres polluted primarily with sulfur dioxide, and that effects increase in seriousness as the atmospheres become more contaminated with other pollutants, such as particulates and sulfuric acid. The more frequent findings include: abnormal urinary acidity; a tendency to increased fatigue; dyspnea; reduced expiratory flow rates; upper respiratory irritations with consequent alteration in sense of smell and taste; nasopharyngitis and bronchitis; and increased incidences of cough and expectoration. There are increased incidences and increased durations of certain respiratory diseases from infections, including colds and pneumonia.

Experimental Exposures of Humans to Sulfur Oxides

Experimental exposures of humans to oxides of sulfur generally have been concentrated on measuring pulmonary function, sensory responses, determining taste and odor thresholds, and studying eye irritation in very brief exposures.

Sim and Pattle[239] exposed healthy males aged 18 to 45 years to sulfur dioxide in concentrations ranging from 1 to 80 ppm for 10 to 60 minutes. With sulfur dioxide dosages below 800, expressed as the product of mg/m^3 and minutes of exposure, or with dosages below 270, expressed as the product of ppm and minutes of exposure, (e.g., 4 to 5 ppm for 1 hour) little change was noted either clinically or in airway resistance as measured by the Ainsworth interrupter. However, lung resistance to airflow occasionally increased by more than 20 percent, and these increases were accompanied by signs of irritation as evidenced by ausculatory indications of moist rales.

In the same investigation subjects were exposed to 4N and 10N sulfuric acid mists ranging from 1 to 10 mg/m^3 of 1 to 1.5μ size for 10 to 80 minutes. Subjects exposed to 10N sulfuric acid mist dosages (mg/m^3 times minutes of exposure) ranging from 41 to 1,230 (e.g., 4 mg/m^3 for 10 min or 20 mg/m^3 for 60 min) failed to show any significant physiological response. However, it was demonstrated that wet mists of lower normality (4N) and larger particle size (1.54μ mass median diameter) were more irritating than dry mists of higher normality (10N) and smaller particle size (0.99μ mass median diameter). The clinical findings were similar to those found for the men exposed to sulfur dioxide at higher concentrations. In most cases lung resistance was increased by more than 20 percent.

Both Sim and Pattle, who were frequently exposed to sulfur dioxide and sulfuric acid mist over the 10 months of the investigation, developed increasing sensitivities to sulfur dioxide and to sulfuric acid which were manifested by persistent bronchitis and which exacerbated to periods of coughing and wheezing upon exposure to either one of these substances. In one investigator persistent wheezing lasted for 4 days following exposure.

Amdur et al.[240] reported changes in respiration and pulse rates in 14 healthy men aged 23 to 58 years who were exposed for 10 minutes to low concentrations of sulfur dioxide (1 to 8 ppm). Lawther[241] could not confirm this in experiments with 18 men aged 21 to 40 years who had no history of cardiopulmonary disease. However, two of Lawther's subjects developed bronchospasm when exposed to 10 ppm of sulfur dioxide and it should be noted that in his experiments measurements of basal pulse and respiration rates and tidal volumes in the control group were consistently higher than during the period corresponding to recovery in the exposed subjects. The question arises as to whether Lawther's subjects were in basal conditions when the measurements were made. If they were not the differences in interpretation of the two studies may be accounted for.

Nadel et al.[242] reported that airway resistance determined with a body plethysmograph in persons exposed to 2 ppm and 5 ppm of sulfur dioxide for 3 to 10 minutes was increased. One individual out of seven exposed to 5 ppm of sulfur dioxide for 10 minutes showed a marked increase in airway resistance which was reversible by isoproterenol inhalation. This subject had no previous history of pulmonary disease.

Frank et al.[243] used a volume-displacement body plethysmograph to study airway resistance, pulmonary compliance, tidal volume, and respiration and pulse rates in 11 healthy volunteers 22 to 56 years of age. They observed increased airway resistance at 5 ppm but not at 1 ppm during 30-minute exposures to sulfur dioxide. The increased resistance developed during the first 5 minutes and was maintained throughout the exposure period. There were no consistent changes in pulmonary compliance, tidal volume, respiration, or pulse rate. However, in measurements taken prior to exposure to sulfur dioxide respiration rate averaged 20 breaths per minute, pulse rate averaged 84 beats per minute and the tidal volume averaged 680 ml, and the question again arises as to whether the subjects were in true basal conditions when measured. Nadel et al.[200] also observed increased airway resistance during 10-minute exposures of 7 healthy adults 27 to 40 years old to 4 to 6 ppm of sulfur dioxide. It was demonstrated that the response could be inhibited by subcutaneous injection of 1.2 to 1.8 mg of atropine sulfate.

Tomono[244] observed that the lowest level of sulfur dioxide which could induce bronchoconstriction, during 10 minute exposures, among 46 healthy male subjects was 1.6 ppm and that this could be relieved by inhalation of isoproterenol hydrochloride.

Amdur et al.[245] and Morando[246] observed shallower and more rapid breathing in subjects inhaling sulfuric acid mist at a concentration of 350 $\mu g/m^3$ in air; a level below the threshold of sensory detection.

Wright[247] observed no changes in airway resistance, maximum and minimum mid-expiratory flow rates, maximum breathing capacity, timed vital capacity, pulmonary volumina, or ventilation effectiveness in normal subjects 20 minutes after exposure to mixtures containing 2.5 to 23 ppm of sulfur dioxide, water aerosol, and 43 to 214 million solid particles (less than 10μ in size) per cubic meter. Airway resistance of persons with emphysema decreased 20 minutes after exposure to mixtures containing 5 ppm of sulfur dioxide, 214 million solid particles per cubic meter, and water aerosol.

Frank et al.[248] studied 12 healthy male volunteers 25 to 56 years old. They observed that increased pulmonary flow resistance measured in the body plethysmograph was the same for exposures to 4 to 17 ppm sulfur dioxide alone as for exposures to sulfur dioxide plus 18 mg/m^3 of sodium chloride aerosol of approximately 0.1μ geometric mean diameter. No increase in airway resistance was demonstrable for exposures to 1 to 2 ppm of sulfur dioxide either with or without salt aerosol. Nakamura[249] observed greater increases in airway resistance from exposure of 10 subjects to 9 to 60 ppm of sulfur dioxide mixed with 0.95μ count median diameter size particles of sodium chloride than from exposure to sulfur dioxide alone. Toyama[250] observed greater increases in pulmonary resistance during 5 minute exposures to 1.5 to 80 ppm of sulfur dioxide mixed with sodium chloride aerosol of 0.22μ count median diameter than during exposures to sulfur dioxide alone. Toyama and

Nakamura[251] studied the effects on airway resistance in 24 healthy male volunteers exposed for 5 minutes with and without bronchodilator to hydrogen peroxide aerosols of 4.6 and 1.8μ count median diameter together with 1.5 to 60 ppm of sulfur dioxide, and of combinations of hydrogen peroxide aerosols and sulfur dioxide. He demonstrated that the aerosols had no effect by themselves at a concentration of approximately 0.3 mg/m^3 but that they augmented the response to sulfur dioxide, that the augmentation was greater with the large particles, and that in all cases the increased airway resistance could be alleviated by bronchodilators. In the simultaneous exposures to sulfur dioxide and hydrogen peroxide some of the sulfur dioxide was oxidized to sulfuric acid to the extent that from 0.01 to 1.4 mg/m^3 of acid existed in the exposure atmosphere. Both Nakamura and Toyama used the airflow interruption technique to determine airway resistance.

Toyama[80] exposed 10 healthy male subjects, aged 20 to 35 years, to dust collected in deposit gauges. The aerosol was generated by a Dautrebande apparatus. The particulate size in inhaled air was 0.5 to 10μ (average 2.0) and its concentration was 10 or 50 μg/m^3. After they inhaled 5 to 10 breaths of the dust suspension, airway resistance in the subjects increased approximately 20 percent. Simultaneous inhalation of sulfur dioxide with dust resulted in greater airway resistance than was produced by either substance alone. Again, the increased airway resistance was alleviated by isoproterenol inhalation.

Of the several investigators[252-256] who have studied ambient concentrations of sulfur oxides in complex mixtures and the effects of the sulfurous pollutants on the eye, none has found any indication that the oxides of sulfur in concentrations found in urban atmospheres have eye-irritating properties.

Cralley[257] exposed human volunteers to 10 to 55 ppm of sulfur dioxide for 30 minutes and 60 minutes and measured the effects on the rate of removal of mucus from the upper respiratory tract. Cralley found that the rate of removal at 10 to 15 ppm of sulfur dioxide was reduced 10 to 15 percent. How the rate of removal of mucus from the upper respiratory tract is affected by sulfur dioxide concentrations below 10 ppm or by longer than 1 hour was not determined. However, the author postulated that exposure to small amounts of irritant gases for long periods of time may lead to chronic irritation of the mucosa, which would affect ciliary activity.

Studies of Sensory Threshold Concentrations

Many recent investigations in Russia have been directed toward determining sulfur oxides threshold concentrations for various sensory responses. These investigations have included determination of odor thresholds and the effects of sulfur dioxide on optical chronaxy, sensitivity of the dark-adapted eye to light, interruption of the alpha (α) rhythm in electroencephalograms, and interference with cortical conditioned reflexes as shown by electroencephalograms. Most of these investigations have been summarized and the more recent methodology described by Ryazanov.[258]

An odor threshold is typically determined in a well-ventilated chamber containing a double-barreled apparatus through which pass two small streams of gas, one a stream of pure air and the other a stream of the test gas. The subject sits in front of the apparatus, sniffs both barrels, and points out the odorous one. This experiment with the same concentration of test gas is repeated over a period of several days. Then the experiment is done with lower concentrations, and the study is continued until the subject, in the majority of instances, denies the presence of an odor or gives erroneous answers. The threshold concentration for the most sensitive subject in a group of volunteers is defined as the threshold for odor perception.[258]

Using the double-barrel apparatus described above, Dubrovskaya[259] conducted sulfur dioxide odor perception threshold tests on 12 subjects, most of whom were at an age at which odor perception was presumed to be most sensitive. Sulfur dioxide concentrations of 0.5 to 13 mg/m^3 (0.16 to 4.3 ppm) were used in 530 threshold determinations. Six test subjects sensed the odor of sulfur dioxide in the range 2.6 to 3.0 mg/m^3; four subjects sensed the odor in the range 1.6 to 2.0 mg/m^3; one sensed the odor in the range 2.1 to 2.5 mg/m^3; and one sensed the odor in the range 3.1 to 3.6 mg/m^3. Thus, the average sulfur dioxide odor threshold concentration for these sensitive persons was 0.8 to 1 ppm and for the more sensitive of these sensitive persons it was 0.5 to 0.7 ppm.

Bushtueva[29] reported that among 10 test subjects the minimum concentration of sulfuric acid aerosol (particle size not given) which was sensed by odor ranged from 0.6 to 0.85 mg/m^3 (average 0.72 mg/m^3).

Popov et al.[260] used an apparatus in which the sulfur dioxide concentration could be changed rapidly. They determined that the odor of sulfur dioxide could be detected at concentrations of 4 mg/m^3 (1.3 ppm). At 4.0 to 6.5 mg/m^3 (1.3 to 2.2 ppm), the majority of their test subjects perceived the gas as a strong odor; a few perceived it as a faint odor. Subjects described concentrations of sulfur dioxide above 8.5 mg/m^3 (2.8 ppm) as having a very sharp odor. The number of subjects involved in these studies was not stated.

The sensitivity of the eye to light while a subject is in darkness increases with time. Several investigations have been made on the effects of inhalation of sulfur oxides on this sensitivity. Typically, measurements of a subject's normal sensitivity are taken in a dark, well-ventilated chamber in complete silence (sudden stimuli, including noise, may change the sensitivity). Each subject is tested once daily following preliminary disadaptation. Light sensitivity is measured at 5- or 10-minute intervals, and a normal curve of increasing sensitivity to light is established from measurements taken over a period of 7 to 10 days.[258]

Dubrovskaya[259] studied the effect of inhaling sulfur dioxide in concentrations from 0.96 to 19.2 mg/m^3 for fifteen minutes before measuring light sensitivity during dark adaptation. The investigator reported that light sensitivity was increased by sulfur dioxide concentrations of 0.96 to 1.8 mg/m^3 (0.32 to 0.6 ppm), that the increase in sensitivity reached a maximum at concentrations of 3.6 to 4.8 mg/m^3 (1.2 to 5 ppm), and that further increases in the sulfur dioxide concentration resulted in progressive lowering of eye sensitivity to light, until at 19.2 mg/m^3 the sensitivity was normal.

In exposures during light adaptation, sulfur dioxide concentrations of 0.6 to 7.2 mg/m^3 (0.2 to 2.4 ppm) caused slight increase in eye sensitivity. Maximum sensitivity was attained at 1.5 mg/m^3 (0.5 ppm); at higher conentrations the increased sensitivity began to abate. Two human subjects were used in these experiments. The odor threshold was between 2.5 and 3.0 mg/m^3 for one subject and between 3.0 and 3.6 mg/m^3 for the other. Thus, changes in sensitivity to light during dark adaptation were caused by sulfur dioxide concentrations below the odor threshold.

Bushtueva[29] studied the effect of sulfuric acid mist on two test subjects sensitivity to light. The test periods lasted 60, 90, and 120 minutes. During the first half-hour sensitivity was measured every 5 minutes, and after that every 10 minutes. In each subject a control curve was established by seven repeated tests, and then the effect on light sensitivity of sulfuric acid aerosol exposure for 4 minutes and for 9 minutes at the 15th and 60th minutes, respectively, was determined. With sulfuric acid mist of undetermined particle size at a concentration of

0.6 mg/m^3 a negligible increase in light sensitivity was detected as a result of the exposure at the 15th minute, but no detectable effect was observed as a result of the exposure at the 60th minute. Concentrations in the range of 0.7 to 0.96 mg/m^3 brought about a well-defined increase in light sensitivity. With 2.4 mg/m^3 increased sensitivity to light was elicited by the exposures at both the 15th and 60th minutes of the test; normal sensitivity was restored in 40 to 50 minutes.

Bushtueva[261] determined in tests made on five subjects that a combination of sulfur dioxide at 1 mg/m^3 (0.35 ppm) and sulfuric acid mist at 0.4 mg/m^3 was below the odor threshold. She then studied the effect of sulfur dioxide, sulfuric acid mist and combinations of the two on sensitivity of the eye to light in three of the subjects. The combination of sulfur dioxide at 0.65 mg/m^3 (0.22 ppm) with sulfuric acid mist at 0.3 mg/m^3 resulted in no change in sensitivity of the eye to light. Approximately a 25 percent increase in light sensitivity resulted from either sulfur dioxide at 3 mg/m^3 (1 ppm) or sulfuric acid mist at 0.7 mg/m^3. The combination of sulfur dioxide at 3 mg/m^3 (1 ppm) with sulfuric acid mist at 0.7 mg/m^3 resulted in approximately a 60 percent increase in light sensitivity. Exposures lasted for 4.5 minutes.

The electroencephalogram is a composite record of the difference in electrical potential between two points, one of which, located on the scalp, is influenced by the potential of large numbers of brain cells located under the recording electrode. The other point can be another point on the scalp or a reference point which is less influenced by brain activity, either because of distance from the brain or because of averaging of several points of different potential. Variations occur with age, and the state of wakefulness and attentiveness, or as a result of incoming sensory stimuli from exteroceptive or interoceptive receptors. In the adult at different points over the scalp the electroencephalogram characteristically shows a fairly uniform frequency from 8 to 12 cycles per second. The dominant frequency, α, is inhibited by eye opening and by mental activity.[262]

Subjects with well-defined α-rhythms studied in an electrically shielded chamber and silence show a temporary desynchronization of the α-rhythm each time they are given a light signal. When the light is excluded, the α-rhythm returns to normal. A concentration of test gas is determined which is so low that by itself it does not cause desynchronization of the α-rhythm. After a subject breathes the gas at this concentration, he receives the light signal. After exposure to this sequence (gas then light) several times (5 to 30 times in 1 day), a subject will show desynchronization before the light; that is, in response to the unperceived odor. Thus, the unperceived odor becomes the conditioning stimulus and brings about the so-called conditioned electrocortical reflex.[258]

Bushtueva et al.[263] reported that 20 second exposures of six human subjects to sulfur dioxide concentration from 0.9 to 3 mg/m^3

(0.3 to 1.2 ppm) produced desynchronization of the α-wave lasting 2 to 6 seconds; at concentrations of 3.0 to 5.0 mg/m^3 (1.2 to 1.9 ppm) desynchronization lasted throughout the 20-second exposure. Exposures to 0.6 mg/m^3 (0.2 ppm) did not cause desynchronization of the α-wave. Exposures to sulfuric acid mist at 0.6 to 0.75 mg/m^3 caused desynchronization of the α-wave, whereas exposures to 0.4 to 0.5 mg/m^3 did not. Thus, the threshold for desynchronization of the α-wave is the same for both substances as the odor threshold or the threshold of irritation of the respiratory tract. In other experiments, Bushtueva demonstrated that electrocortical conditioned reflexes could be developed with sulfur dioxide at 0.6 mg/m^3 (0.2 ppm) or with sulfuric acid mist at 0.4 mg/m^3, but not with lesser concentrations of either substance. Finally, Bushtueva[264] demonstrated that combinations of sulfur dioxide at 0.50 mg/m^3 (0.17 ppm) with sulfuric acid mist at 0.15 mg/m^3 or sulfur dioxide at 0.25 mg/m^3 (0.08 ppm) with sulfuric acid mist at 0.30 mg/m^3 could produce electrocortical conditioned reflexes.

Chronaxy is defined as the time required for excitation of a nervous element by a definite stimulus. In the determination of optical chronaxy a weak electrical current is applied to the eyeball to give the sensation of a light flash. For each subject there is an intensity of stimulation (measured in volts) below which no sensation of light takes place. The time required for this minimal voltage to produce the sensation of light in a subject is the optical chronaxy for the subject. According to

Pavlovian theory, the excitation of one area of the cerebral cortex may inhibit the excitation of other areas through the rule of induction.[258] Therefore, it has been postulated that excitation of the olfactory sensory area by the oxides of sulfur inhibits the light-sensing area of the cerebral cortex and thus increases optical chronaxy.

Bushtueva[261] studied the effects of different concentrations of sulfur dioxide, sulfuric acid mist, and combinations of the two on the optical chronaxy of three subjects. Optical chronaxy was determined in each test subject at 3-minute intervals as follows: at the start and on the 3rd, 6th, 9th, 12th and 15th minutes. Between the 6th and 9th minutes the subjects inhaled sulfur dioxide, sulfuric acid mist, or their combination for 2 minutes. In each subject the threshold concentrations of sulfur dioxide and sulfuric acid mist were first determined independently, and then threshold concentrations for combinations of the two were determined.

Results presented for one subject were: (1) neither sulfur dioxide at 0.3 ppm nor sulfuric acid mist at 600 $\mu g/m^3$ produced increased optical chronaxy, but the combination in these concentrations produced a 16 percent increase in optical chronaxy; (2) concentrations of sulfur dioxide at 0.4 ppm with sulfuric acid mist at 400 $\mu g/m^3$ produced no increase in optical chronaxy; and (3) concentrations of either sulfur dioxide at 0.5 ppm or sulfuric acid mist at 750 $\mu g/m^3$ increased optical chronaxy, and the effects of the combination at these concentrations were additive. Similar results were reported for the other subjects.

The data obtained by Bushtueva and her co-workers are summarized in table 8.

The practical ramifications of these neurophysiological responses have not been explored. Certainly, unpleasant odors are not desirable, but changes in sensitivity to light of the dark-adapted eye and impaired transmission of nerve impulses could have more serious consequences, not the least of which would be increased accident rates.

Clinical-Epidemiological Studies on Humans in Community Atmospheres

Clinical-epidemiological studies on the acute and chronic effects of community atmospheres containing oxides of sulfur at various concentrations have been undertaken by numerous investigators. It should be noted that these atmospheres contained other pollutants as well, and although sulfur dioxide is the index of pollution in the studies, also present were undetermined amounts of sulfuric acid mist and sulfate salts as well as other pollutants which could have contributed to the observed effects. It has been shown by numerous investigators that the effects related to a given amount of sulfur dioxide in community air can be expected to be greater than the effects resulting from laboratory experiments in which sulfur dioxide was essentially the only pollutant.

Schoettlin[265] studied 400 men aged 45 to 74 years, 200 of whom had chronic respiratory symptoms and 200 who had normal respiratory function. Age and smoking habits were taken into account. The study was conducted in Los Angeles between August 12

Table 8. Threshold Concentrations of Sulfur Dioxide, Sulfuric Acid and Their Combinations for Various Reflex Responses. Bushtueva[264]

Procedure used	Threshold concentration			
	H_2SO_4	SO_2*	$H_2SO_4+SO_2$	
	μg/m^3	ppm	μg/m^3	ppm
Threshold concentration of irritation effects and odor perception	600-850	0.5-0.9	300	0.17
Data obtained by the method of eye adaptation to darkness	630-730	0.31	300	0.17
Data obtained by the method of optical chronaxy	730	0.50	600	0.40
Encephalographic method	630	0.30	300	0.17
"Electrocortical" conditioned reflex	400	0.20	150 300	0.17 0.08

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* A factor of 0.33 has been used throughout the report to convert mg/m^3 to ppm.

and December 18, 1958. It consisted of evaluating symptoms of chronic respiratory disease and taking pulmonary function measurements such as Puffmeter readings, timed vital capacity, and total vital capacity and then relating the data to air pollution measurements that included sulfur dioxide concentrations. The concentrations during the period of the study were not reported. Multiple linear regression analysis revealed no statistically significant correlation coefficients, but variation in sulfur dioxide and oxidant precursor concentrations accounted for 24 and 30 percent respectively of the variation in symptoms of chronic respiratory disease in the diseased group.

Carey et al.[266-268] studied timed vital capacity and carbon monoxide diffusion rates (a measure of the effectiveness of gas transport across the alveolar membrane) in 10 patients with severe cardiorespiratory disease in Cincinnati. They observed minimal responses to changes in air pollution, which included an average of 0.03 ppm sulfur dioxide with peaks up to 0.25 ppm. However, multiple regression analysis suggested that in some patients significant changes occurred in relation to combinations of smoke, temperature, and humidity. Also, it may be that the patients did have some bronchoconstriction (evidenced by symptoms of wheezing), but that they had recovered from this transient effect before timed vital capacity tests were carried out.

Over a period of several weeks, Spicer[269-270] made daily observations of air pollution and pulmonary function of a group of normal individuals and a group of subjects with chronic obstructive airway diseases. In general, the airway resistance and other measures of pulmonary function of the two groups changed together from week to week, and persons within either group changed together from day to day. Sulfur dioxide concentrations averaged over 2-hour periods ranged from close to zero up to 0.5 ppm during the period of the study.

Lawther and Waller[271-275] and Pemberton[276] studied groups of patients with chronic bronchitis over several winters. Lawther's group consisted of about 1,000 patients with established bronchitis who recorded daily whether they felt better or worse than on the preceding day. The percentage of those who felt worse than on the preceding day was then calculated. Fluctuations in this index followed the daily variations in air pollution closely, and when smoke rose above 300 $\mu g/m^3$ and sulfur dioxide above 0.21 ppm the health of the group deteriorated.[319] The authors concluded that air pollution as measured by smoke and sulfur dioxide was the most important factor affecting the day-to-day well-being of the subjects.

McCarroll et al.[277-281] studied a large number of families (approximately 1,000 people) in New York City and correlated the symptoms of upper respiratory tract and eye irritation with sulfur dioxide concentrations. The incidences of rhinitis, sore throat, cough, and eye irritation increased significantly during two air pollution episodes in the period November through December of 1963 when the daily average sulfur dioxide concentration exceeded 0.2 ppm for 4 days and reached maximum 1 to 2-hour average concentrations of 0.9 to 1.5 ppm.

Motley et al.[282] studied patients in Los Angeles with advanced pulmonary emphysema. They found that the increased residual volume of the patients could be significantly reduced if they were placed for 40 or more hours in a room whose air supply was filtered through charcoal. This was not true for individuals who were not afflicted with chronic obstructive airway disease. Aerometric studies of Los Angeles air prior to filtration revealed maximum daily concentrations as follows: total oxidants 0.2 to 0.7 ppm (ozone 0.2 to 0.5 ppm); oxides of nitrogen 0.2 to 0.5 ppm; sulfur dioxide 0.05 to 0.25 ppm; and carbon monoxide 5 to 27 ppm. No measurements of pollutants in the air were made after filtration. No effort was made to determine the role of each specific pollutant.

After controlling for age, height, sex, and smoking habits, Anderson and Ferris[283] found that 1-second forced expiratory volume and Wright peak expiratory flow rates were greater for persons living in a Canadian town with less pollution (0.05 mg SO_3/100 cm^2/day; 10 tons/mi^2/mo dustfall) than for persons living in a more polluted United States town (0.4 mg SO_3/100 cm^2/day; 35 tons/mi^2/mo dustfall). In both towns air pollution was measured only during August and September, months of generally low pollution in the United States and Canada. Factors other than air pollution such as ethnic group may account for the differences in the pulmonary function measurements.

Toyama,[80] compared two groups of primary school children, one living in a relatively polluted area and the other living in a relatively unpolluted area. He found significant correlations of monthly dustfall and lead peroxide candle sulfation rates with measurements of respiratory flow rates from the Wright peak flow meter. Children residing in the more polluted area were more often found to have non-productive cough, a mucous membrane irritation of the upper respiratory tract, and increased mucous secretion. Total vital capicity measurements in the two groups were similar. Monthly dustfall was 18 to 36 tons/mi^2/mo in the less polluted area and 40 to 182 tons/mi^2/mo in the more polluted area. Lead peroxide candle sulfation rates from month to month ranged from 0.5 to 2 mg SO_3/100 cm^2/day in the higher polluted area (20 month average 1.2 mg SO_3/100 cm^2/day). For the purpose of comparison with the data of Anderson and Ferris,[283] it is noted that the lowest 3-month average during the year was 0.67 mg SO_3/100 cm^2/day.

Prindle et al.[284] observed greater airway resistance in persons who lived in the more heavily polluted of two towns which were located about 5 miles apart. Factors other than air pollution such as smoking habits were not taken into consideration. Sulfation rates (3.7 mg SO_3/100 cm^2/day) and dustfall 83 tons/mi^2/mo) were respectively 6.2 and 3.2 times greater in the one town than in the other. Gaseous sulfur dioxide as indicated by the West-Gaeke technique averaged 0.09 and 0.01 ppm from October 6, 1959 to April 26, 1960 in the town with high air pollution concentrations and the one with the low concentrations, respectively. The major source of pollution was a coal-fired power plant located a half-mile from the more polluted town.

Bell[285] found no significant difference in 1-second forced expiratory volume between persons living in a more heavily polluted area and those living in a cleaner area of Port Kembla, New South Wales, Australia, although he did observe higher incidences of bronchitis, nasal catarrh, head colds, wheezing, chronic cough, and increased phlegm among persons living in the more polluted area. Smoking habits and occupational histories were similar in the two areas. Sulfur dioxide concentrations were higher during the summer than during the winter, so that there was no question of whether the effects were due to high pollution or low temperature, as has been the case in other countries. The average concentrations indicated by the hydrogen peroxide technique for the two areas were 0.034 ppm for the more polluted area and 0.009 ppm for the cleaner area. However, since the pollution was mainly from a few large sources (steel mills, copper smelters, and power plants), high concentrations occurred occasionally for brief periods. Measured by the hydrogen peroxide technique daily average concentrations exceeded 0.2 ppm at the most polluted air monitoring station on approximately 3 percent of the days. Measured with the Thomas Autometer the highest concentration at this station lasting for several minutes was 13.5 ppm, and peak concentrations reached 1 to 5 ppm on several occasions. Cohs values in the more polluted and the cleaner area averaged over 1 year were 2.7 and 1.3 respectively.

Elfimova and Shashkov[286] measured sulfur dioxide concentrations between 0.01 and 0.02 mg percent in the blood of 17 of 62 subjects who lived in a highly polluted area where the average ambient sulfur dioxide content was 0.7 ppm, but no more than traces in the blood of any of 30 persons who lived in an area where the average ambient sulfur dioxide concentration was 0.3 ppm.

Yanysheva[287] found that respiratory diseases, anemia, and rickets among children living in a highly polluted area (4.4 to 10 ppm sulfur dioxide) were 1.6 to 3.8 times higher than in children in a control area of the same city. Lung x-rays showed fibrotic changes in a higher percentage of the children from the more polluted area -- 14.6 per cent compared to 4.2 percent.

Epidemiologic Studies of Morbidity Within Cities

Diseases which seem to be of the most concern from the standpoint of sulfur oxides pollution are chronic bronchitis and/or chronic disabling respiratory diseases, cardiac diseases, and respiratory diseases due to infection.[288-292]

Martin[293] studied daily hospital admissions in London during the periods 1958 to 1959 and 1959 to 1960. The hospital admissions were separated into three categories: (1) diseases from all causes; (2) cardiac diseases, and (3) respiratory diseases. For each category a daily morbidity index was determined as the deviation from the 15-day moving average of the number of admissions. The separate daily morbidity indexes were than correlated with daily measures of smoke and sulfur dioxide. Correlation coefficients of morbidity from all causes with smoke and sulfur dioxide were significant only in 1958 to 1959. The correlation coefficients of respiratory morbidity with smoke and with sulfur dioxide were both approximately 0.25 in 1958 to 1959 and were both approximately 0.34 in 1959 to 1960. Correlation coefficients of cardiac diseases with smoke were 0.28 in 1958 to 1959 and 0.22 in 1959 to 1960; those of cardiac disease with sulfur dioxide were 0.20 in 1958 to 1959 and 0.23 in 1959 to 1960.

Burn and Pemberton[294] observed that in Salford, England, during 1958, up to four times as many bronchitis attacks were reported as were expected at that time of the year during five smog periods. Daily average sulfur dioxide concentrations during these periods were between 0.5 and 1 ppm, and daily average smoke concentrations were above 1 mg/m^3. During the year period, bronchitis attacks ranged from 130 percent of expectation in the most severely polluted area to 60 percent in the least polluted area. Average daily sulfur dioxide concentrations during the winter months in the more heavily polluted areas were approximately 0.25 ppm; in the less polluted areas corresponding average daily sulfur dioxide concentrations were approximately 0.10 ppm. Average daily smoke values were approximately 500 $\mu g/m^3$ in the more severely polluted areas and 350 $\mu g/m^3$ in the less polluted areas.

Ferris and co-workers[295-296] surveyed Berlin, New Hampshire and found no consistent differences in the prevalence of respiratory diseases in greater and lesser polluted areas. In the most heavily polluted area during August and September, sulfur dioxide averaged 0.02 ppm, dustfall 46.7 tons/mi^2/mo, and sulfate in dustfall 10.6 tons/mi^2/mo.

In a Detroit, Michigan/Windsor, Ontario, study, greater morbidity was observed in areas with higher pollution than in areas with lower pollution.[297] Sulfur dioxide concentrations were about twice as high in the more polluted areas (0.04 to 0.10 ppm annual average) as in the lesser polluted areas. In the more highly polluted areas suspended particulates were found to be 1.2 to 1.8 times greater (193 to 281 $\mu g/m^3$ annual average) than in the lesser polluted areas. As absolute differences in pollution between otherwise comparable areas increased, greater differences in sickness rate between the areas were noted.

Zeidberg et al.[298] studied morbidity among 9,313 individuals in 2,833 households in Nashville, Tennessee. The major part of the analysis was confined to the middle socioeconomic class households representing 6,393 individuals. No direct correlation between morbidity and pollution could be shown with any consistency except for those 55 years of age or older and for housekeeping females. Cardiovascular morbidity among the white population in the older age group and total illness rates among the housekeeping females progressed as either the soiling index or sulfur dioxide concentration in the residential area increased. Cardiovascular morbidity among the white population 55 years of age or older was approximately twice as high in the most polluted areas (annual average--Cohs 0.831 or more, sulfur dioxide 0.01 ppm or more) as in the least polluted areas (Cohs 0.330 or less, sulfur dioxide 0.005 ppm or less). The annual averages are geometric means which can be approximately converted to arithmetic means by multiplying by 1.5. Relationships between air pollution and morbidity rates for cancer, respiratory diseases, and gastrointestinal disease could not be demonstrated.

Sterling et al.[299] studied daily admissions of members of Blue Cross groups to hospitals of 100 beds or more which were within 5 miles of an air pollution monitoring station. The period of the study was from March 17, 1961 to October 26, 1961. Diseases were grouped with relevancy to air pollution as: Highly Relevant (allergic disorders, inflammatory diseases of the eye, acute upper respiratory infections, influenza and bronchitis); Relevant (diseases of the heart, rheumatic fever and vascular diseases, other diseases of the respiratory systems); and Irrelevant (all other illnesses). Statistically significant correlations existed between the highly relevant diseases and 9 air pollutants. Statistically significant correlations with relevant diseases existed only with oxidant, ozone and sulfur dioxide. A detailed analysis of the relationship of hospital admissions on the days of highest and lowest sulfur dioxide concentrations indicated increased admissions on the highest days from infectious diseases; malignant neoplasms of the respiratory system; endocrine, metabolic and nutritional diseases; rheumatic fever and vascular diseases; acute upper respiratory infections; bronchitis; disease of tonsils and adenoids; and other respiratory diseases. Sulfur dioxide concentrations during the period of the study averaged less than 0.015 ppm. Concentrations on days of highest pollution were not reported.

Petrilli et al.[300] studied the frequency of respiratory symptoms (determined by use of a modified British Medical Research Council questionaire) in non-smoking women over 65 years of age who had lived in their respective areas for a long time and who had never worked in industry. The frequencies of cough, sputum, dyspnea, and bronchitis were all greater in an area with annual averages of 0.037 ppm sulfur dioxide and 185 $\mu g/m^3$ of suspended particulate matter than in an area with 0.028 ppm sulfur dioxide and 90 $\mu g/m^3$ of suspended particulates. They also studied morbidity in persons under daily medical observation. Annual morbidity indices were calculated for groups exposed to different air pollution levels, but considered homogeneous from the viewpoint of economic and social conditions. The correlation coefficient between bronchitis morbidity and sulfur dioxide concentrations was 0.98. The correlations with suspended matter (0.82) and dustfall (0.66) were not significant. Multiple correlation analysis indicated that sulfur dioxide was much more important than winter temperature in determining bronchitis morbidity.

Epidemiologic Studies of Morbidity Between Cities

Fairbairn and Reid[301] used the percent of days during a year on which the visibility at 9:00 a.m. was 100 yards or less as an index of air pollution, and found a high correlation between this index and "bronchitis wastage" (man-years lost due to death in service or early retirement as a result of bronchitis) in postmen. They compared the morbidity of indoor workers with that of outdoor workers and found: (1) a higher incidence in the outdoor workers of upper respiratory tract infections, such as colds, sore throats, and influenza; and (2) an earlier onset of bronchitis among outdoor workers.

Dohan[302-303] studied the incidence of respiratory illnesses lasting more than 7 days in female employees in five cities. The results of this study showed a very high correlation (0.964) between the average concentration of suspended sulfate in the air and the rate of respiratory illnesses in these cities. The average sulfate concentrations for the cities ranged from 7.4 to 19.8 $\mu g/m^3$. During the non-influenza epidemic years the incidence of respiratory disease in the city with the highest sulfate concentration was more than twice as great as the incidence in the city with the lowest sulfate concentration.

The incidence of total respiratory disease during the 1957 to 1958 Asian influenza epidemic was greater than in the same cities during non-epidemic years. In the city with the lowest concentration of suspended sulfates there was approximately a 20 percent increase in the incidence of respiratory illness during the epidemic year, whereas in the city with the highest concentration of suspended sulfate there was approximately a 200 percent increase in the incidence of respiratory illness. During the influenza year the incidence of respiratory diseases in the city with the highest pollution was more than 5 times as high as in the city with the least pollution. The author could not demonstrate correlations between respiratory disease rates and the mean concentrations of benzene soluble organic matter, acetone soluble organic matter, nitrates, cooper, or zinc. However, when the mean concentrations of nickel and vanadium (measured in 4 cities) increased the incidence of respiratory disease also increased.

Epidemiologic Studies of Morbidity Associated with Air Pollution Episodes

Increased illness rates associated with the acute air pollution episodes which occurred in the Meuse Valley in 1930, Donora in 1948, New York City in 1953 and 1962, and London in 1952 and 1962, and in which high concentrations of sulfur oxides were present have been reported.[304-314]

In the Meuse Valley, Belgium, during a period of anticyclonic weather in December, 1930, a dense fog enveloped the valley. By the third day many of the residents developed throat irritation, hoarseness, productive and non-productive cough, shortness of breath, and sense of chest constriction. Some individuals also developed nausea and vomiting. The most severely affected were elderly people and individuals who had previous cardiorespiratory disease. No measurements of air pollution were made at the time of the episode, but subsequent investigations indicated that the oxides of sulfur were the principle irritant.[304-305]

In Donora, Pennsylvania, in October, 1948, during a similar meteorological condition which lasted 4 days, 42.7 percent of the population (5,910 persons) developed mild to severe symptoms of irritation characterized by burning of the eyes, tearing, nasal discharge, sore throat, non-productive cough, nausea, vomiting, and diarrhea. No age group was spared, but the incidence rate of illness increased with age. Regardless of age the most sensitive individuals were those with pre-existing heart and lung disease.[306] Retrospective studies indicated that sulfur dioxide levels may have reached 0.5 to 2.0 ppm and that large numbers of other air-borne particulates and gases were present.[307]

Ten years after the incident in Donora, Ciocco and Thompson[308] restudied that part of the population who had lived there during the episode. Among the persons surviving in 1957 who could be questioned (80 percent of the total study group), no evidence showed that those who smoked tobacco in any form prior to October, 1948, became ill during the episode at a higher rate than those who did not smoke. The essential findings were that persons who reported acute illness at the time of the episode subsequently demonstrated higher mortality and higher prevalence of illness than the other persons living in the community at that time.

Abercrombie[309] reported that the normal number of weekly applications for emergency bed service during the month of December in London was approximately 1,000. In 1952 during a period of extremely high air pollution accompanied by fog, the weekly total number of applications between December 5 and 9 was more than 2,500. The increase in illness was largely in cardiorespiratory diseases. The illness rate did not return to the normal statistical rate for approximately 2 to 3 weeks. Sulfur dioxide measurements reached peak levels of 1.3 ppm and the general average during the episode was 0.57 ppm.[335]

Greenburg et al.[310] reported that during the period of high air pollution in New York City in November, 1953, pediatric and adult clinic visits for upper respiratory illnesses and cardiac diseases rose above normal in all of the four hospitals studied. Sulfur dioxide ranged between 0.07 and 0.86 ppm from November 12 to 24, and hospital admissions were clearly elevated by November 16, at which time concentrations had not exceeded 0.25 ppm.

From November 27 through December 4, 1962, a condition of atmospheric stagnation occurred in New York City. Sulfur dioxide concentrations ranged up to 1.4 ppm and Cohs values up to 9 were observed. Greenburg et al.[311] studied visits during this period for upper respiratory infections, cardiac conditions, and asthma at five emergency clinics in the major city hospitals and at an employee clinic at the Chase National Bank; they also studied the incidence of these illnesses at four old age homes and in hospital visits recorded by the Blue Shield Health Insurance Plan. The average number of daily visits at each of the installations were compared with those during the periods prior to and subsequent to December 1 through 7. No significant change was found in the records of any of the facilities except those of the 4 old age homes. A significant rise in upper respiratory illness was found in all 4 of the old age homes, and the incidence of illness did not return to normal until after December 14.

Acute illnesses, of epidemic magnitude have developed after measured 24 hour average sulfur dioxide concentrations reached approximately 0.5 ppm, with peak hourly averages of 0.75 ppm or more, accompanied by suspended particulate matter concentrations of 1,000 $\mu g/m^3$ or higher or Cohs values of 8 or more. Increased hospitalization and outpatient clinic visits were primarily attributable to cardiorespiratory illnesses. In severe episodes accompanied by fog, nausea and vomiting occurred in addition to the usual symptoms of respiratory irritation. Secondary complications frequently developed in individuals of all age groups, but elderly individuals and individuals with pre-existing cardiorespiratory disease were especially susceptible. From one study it was determined 10 years after the episode that individuals who became ill during the outbreak had less favorable morbidity and mortality experiences than those who were not so affected.

Epidemiologic Studies of Mortality Within Cities

Deaths within cities have been studied in relation to air pollution containing oxides of sulfur.[294,315-326] In general, very high correlations of total daily deaths from "all causes" and deaths from bronchitis with daily changes in both sulfur oxides and suspended particulate matter were obtained. The two variables, sulfur oxides and suspended particulate matter, correlate highly with each other and with temperature. Although it is difficult to isolate the relative importance of each, increases in deaths in London from "all causes" and increases in deaths from bronchitis occur within 24 hours after daily average sulfur dioxide concentrations reach about 0.25 ppm and black suspended matter rises above 750 $\mu g/m^3$. These increases may occur at lower concentrations when the daily average sulfur dioxide concentration is 0.025 ppm greater than the average of the previous day.[315]

Burn and Pemberton[294] reported the death rate from bronchitis was approximately twice as high in the high pollution area of Salford, England, as in the low pollution area. In the high pollution area annual average sulfur dioxide was about 0.15 ppm and in the low pollution area it was about 0.08 ppm. The authors point out that although the higher morbidity and mortality rates from bronchitis in the most polluted area are consistent with the hypothesis that air pollution is a cause of incapacity and death from bronchitis, other causes cannot be completely ruled out. They state, for example, that it is possible that people living in the more polluted parts of the city smoke more cigaretts than those living in the cleaner parts; however, figures available on smoking habits (from the Tobacco Manufacturers' Standing Committee, 1959) suggest that differences in smoking habits in different parts of the city are small.

A relationship between bronchitis death rates and degree of pollution in residential areas of London could not be demonstrated.[316]

In Nashville, Tennessee, the total respiratory disease death rate among the middle socioeconomic class in the areas where the annual average lead peroxide candle sulfation rates were 0.4 mg SO_3/100 cm^2/day or more (geometric mean 0.492 mg SO_3/100 cm^2/day), where Cohs were 1.1 or more, or where dustfall was 12 or more tons/mi^2/mo was nearly twice as high as in areas where these pollutants were less concentrated (average sulfation rate 0.189 mg SO_3/100 cm^2/day).[317] These pollution concentrations are expressed as geometric means and can be approximately converted to arithmetic means by multiplying by 1.5.

Epidemiologic Studies of Deaths Among Cities

Studies of differences in death rates as they relate to air pollution among different cities of the United States have not been made. However, several such studies have been made in England.[327-331] Very high correlations have been obtained between various measures of air pollution containing sulfur oxides and respiratory disease deaths. Bronchitis death rates can be closely related to indices of air pollution from domestic coal consumption,[327] to lead peroxide candle sulfation rates,[328] and to pH of the precipitation.[329] Pneumonia death rates increase from 40 per 100,000 to 60 per 100,000 as sulfate in the dustfall increases from 1.4 to 7 tons/mi^2/mo.[330] Bronchitis death rates increase by a factor of about 1.5 as lead peroxide candle sulfation rates increase from 0.75 to 2.25 mg SO_3/100 cm^2/day.[328]

Epidemiologic Studies of Deaths During Air Pollution Episodes

Numerous reports of increased deaths during air pollution episodes exist.[297,304-306,332-347] In London, England, annual average sulfur dioxide concentrations are about 0.10 ppm, but fogs are frequent, and air pollution episodes occur in varying degrees almost every year.[332-344] In the most notable episode, which occurred during December 5 to 9, 1952, there was 4,000 excess deaths. During this episode sulfur dioxide averaged 0.73 ppm in the central area during the 4-day period and reached a maximum daily average of 1.3 ppm. For the entire London area, sulfur dioxide measured at 12 sites averaged 0.57 ppm for the 5 days and smoke averaged 1.41 mg/m^3. The maximum daily average smoke concentration was 4.46 mg/m^3.[332-337] In the United States only a few incidents of excess deaths during periods with excessive concentrations of air pollution have been recorded, but an intensive search for their occurrence has not been made.

In the 4-day air pollution episode at Donora, Pennsylvania, in October, 1948, 17 persons died on the third day and 3 subsequent deaths were ascribed to conditions which developed during the episode. At Donora in the period 1945 to 1948 about 100 persons died each year or 1 person every third day. The ages of the 20 persons who died during the episode ranged from 52 to 84 years. Pre-existing disease of the cardiorespiratory system appeared as a single factor among the fatally ill, although in four cases no history of any chronic desease was obtained. Autopsies of three persons who died during the smog showed acute changes in the lungs characterized by capillary dilation, hemorrhage, edema, purulent bronchitis, and bronchiolitis. Chronic cardiovascular disease was a prominent feature in the autopsies. Only in the degree of severity and in the outcome were the fatal cases different clinically from the severely ill persons who did not die.[306]

In the air pollution episode that occurred in November, 1953 in New York City, the average number of deaths per day in the period November 15 to 24 was 244, whereas the averages for 6 control years ranged from 218 to 227. The increase was generally distributed over all age groups and all causes of deaths except accidents, homicides, and suicides.[345]

Greenburg et al.[346] studied mortality in New York City from January 29 through February 12, 1963. This was a period of cold weather and high air pollution (sulfur dioxide averaged 0.46 ppm with a peak of 1.5 ppm for 4 hours; Cohs values were 4 or more on 7 days) during which A_2 virus was prevalent in the population. A similar period of cold weather when the A_2 virus was also active, but when air pollution was lower (sulfur dioxide 0.19 ppm) existed on the same dates in 1958. In 1958 deaths averaged 293 per day the week immediately prior to and the week immediately subsequent to the critical period; in 1963 the deaths averaged 283 per day during these control weeks. In 1958 deaths averages 279 per day from January 29 to February 12, whereas in 1963 they averaged 306 per day during this period. The total excess deaths due to cold virus and air pollution were estimated to be 647 and those due to air pollution were estimated to be 406. The increase in mortality took place among the older age groups (45-64 and 65 and over) and for the causes, influenza-pneumonia, vascular lesions, cardiac and "all other". No significant increase in accidental deaths, homicides, suicides and deaths of early infancy was found.

During a 3 day stagnation period in Detroit, in September, 1952, suspended particulates rose by a factor of 2.5 (from a normal of about 210 to 500 $\mu g/m^3$). Sulfur dioxide averaged 0.04 ppm during the month, and if the same relationship between the average concentrations and the episode concentrations can be assumed for sulfur dioxide as for particulates, the maximum daily average during the stagnation period would have been about 0.08 ppm. An instantaneous maximum of 1 ppm was observed. During the stagnation period, fogs were present each morning between 6:30 and 9:00 a.m. During the remainder of the days, the relative humidity was low.[347] Both infant mortality and cancer mortality increased from normal by a factor of about 2 during this 3 day period.[297]

Existing data, then, indicate that excess deaths may occur in United States cities with annual average sulfur dioxide concentrations of 0.05 ppm or more and/or during stagnation periods of 3 or more days in which peak instantaneous concentrations of 1 ppm or 1/2-hour average concentrations of 0.8 ppm occur. Deaths are not limited to any particular group, but certain groups, including those with pre-existing heart, circulatory, or respiratory disease, and infants and the elderly, are unquestionably affected.

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